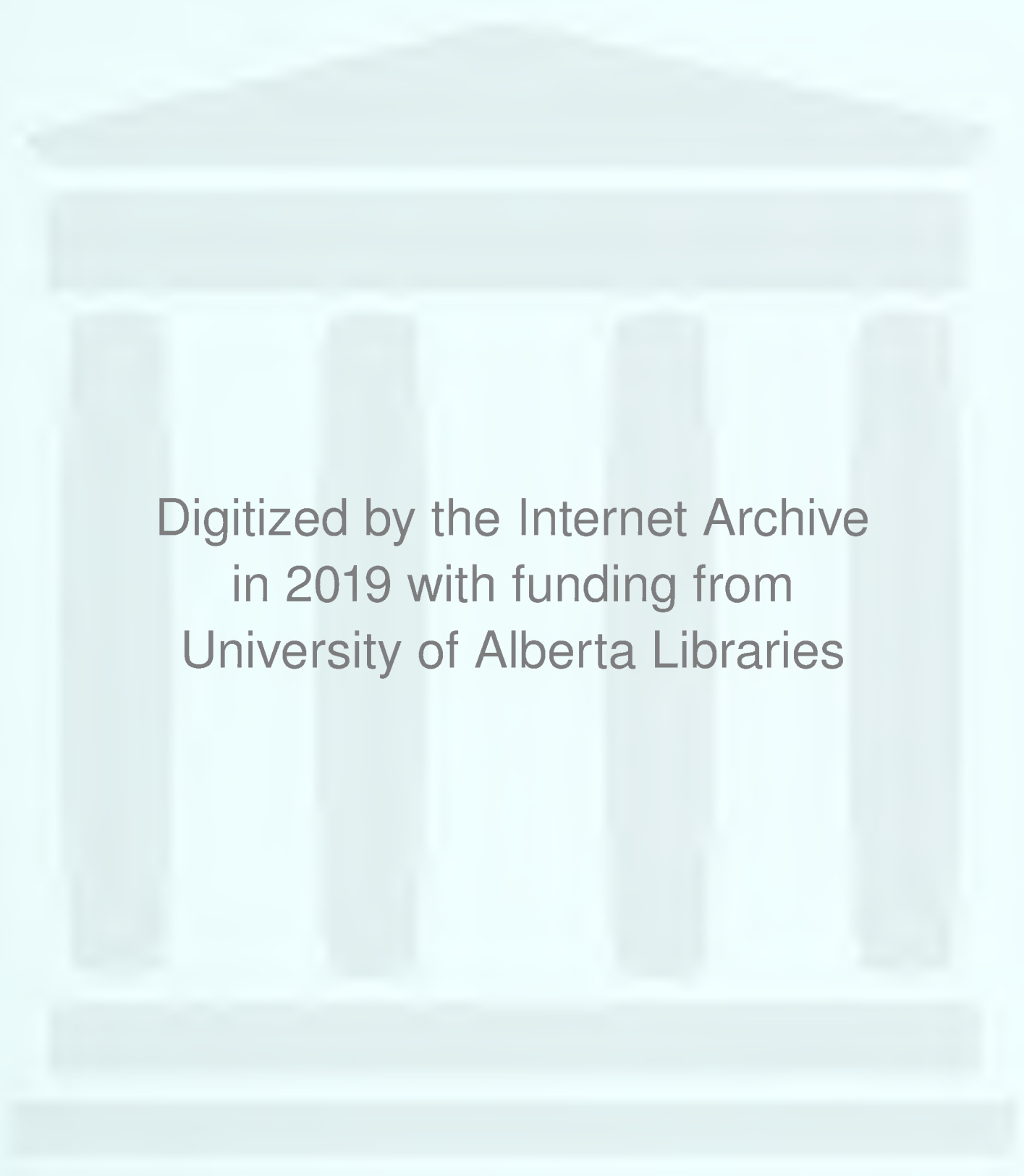




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"LIQUID LIQUID EXTRACTION OF ACETIC ACID
USING COCURRENT FLOW THROUGH A PACKED COLUMN"

by

G.P. Jain

A THESIS

SUBMITTED TO THE FACULTY OF GRADUATE STUDIES
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UNIVERSITY OF ALBERTA
FACULTY OF GRADUATE STUDIES

The undersigned certify that they have read, and recommend to the Faculty of Graduate Studies for acceptance, a thesis entitled "Liquid-Liquid Extraction of Acetic Acid Using Cocurrent Flow Through a Packed Column" submitted by G.P. Jain in partial fulfilment of the requirements for the degree of Master of Science in Chemical Engineering.

ABSTRACT

The purpose of this study was to investigate mass transfer in cocurrent flow in packed beds. In this work extraction of acetic acid from dilute aqueous solutions using benzene and ethyl acetate has been studied.

The principal part of the experimental apparatus was a 1/2-inch I.D. glass column packed with 3 mm glass beads. Individual phase flow rates of approximately 100 to 400 cc/min. were investigated over a complete range of flow ratios. The concentration of acetic acid in either phase was determined by volumetric analysis.

Overall mass transfer coefficients for the benzene-acetic acid-water system were up to 86 times greater than those obtained by other workers in countercurrent flow in packed beds. In the ethyl acetate-acetic acid-water system the overall mass transfer coefficients were up to 130 times greater than those obtained by other workers for countercurrent flow in packed beds. The mass transfer coefficients increased with increasing flow rates and even higher mass transfer coefficients are apparently attainable with higher flow rates. In the benzene-acetic acid-water system bed height had no effect on the overall mass transfer coefficient.

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I. INTRODUCTION

In countercurrent operation the flooding point places an operational limit upon the capacity of the packed column. In contrast, in cocurrent operation, the column has no such limit and the flow rates are limited only by the allowable pressure drop. Higher mass transfer coefficients can be obtained by using higher velocities. The concentration to bed height relationship in cocurrent flow is similar to that in countercurrent flow when the change in concentration of the transferring component is negligible or essentially constant in one phase. Under this condition the cocurrent operation of a packed bed may be comparable to the countercurrent operation.

Mixer settler units are used in liquid-liquid extraction. The two phases are mixed in the mixer by an agitator and the resulting emulsion or dispersion is settled and decanted in the settler. These units are operated in series to obtain more than one equilibrium stage. Mechanical agitators for the mixer are generally bulky, inefficient, and expensive. On the other hand, cocurrent flow through a packed bed might provide an efficient and compact design for inexpensive mixing and dispersion. The power required for dispersion and mixing would be supplied by the pressure drop across the packed bed. Mixer settler units employing packed beds as mixing devices could be operated in series to obtain more than one equilibrium stage.

Leacock and Churchill(9) investigated the mass transfer in cocurrent flow between liquid phases. They studied a two component system and obtained very high mass transfer coefficients. Most of the commercial applications of liquid-liquid extraction involve extraction of a solute from one phase by another immiscible phase. This work was undertaken to study the mass transfer between immiscible liquids in cocurrent flow in packed towers and to determine the important variables and their effect on the overall mass transfer coefficient. In this investigation three component systems were studied and no work has been reported in the literature on a three component system in cocurrent liquid-liquid extraction. In industry acetic acid is recovered by liquid-liquid extraction processes. Thus, in this work extraction of acetic acid from dilute aqueous solutions by benzene and ethyl acetate was studied. The benzene-acetic acid-water system was chosen because a large amount of data in packed beds, wetted wall columns and spray columns was available for comparison. The solvent was readily available and the technique of acetic acid analysis was simple. Below about 16 percent by weight of acetic acid extraction with ethyl acetate has been reported as the most economic method of recovery. Hence, the ethyl acetate-acetic acid-water system was studied to compare the magnitude of mass transfer coefficients in cocurrent flow with the results of other workers using countercurrent flow.

II. LITERATURE REVIEW

An exhaustive survey of periodical literature has shown that no work on cocurrent liquid-liquid extraction through a packed bed in a three-component system has been published. However, there is a large number of papers which are of value to this investigation. The more important papers are reviewed in this section. The supplementary bibliography in Appendix E lists articles which are indirectly related to this study.

1. Mass Transfer in Cocurrent Flow

1.1 Liquid-Liquid Systems

Leacock and Churchill(10) studied mass transfer in the isobutanol-water system at 25°C. The individual mass transfer coefficients were calculated by assuming that the interface concentrations were equal to the static equilibrium concentrations. In cocurrent flow they obtained mass transfer coefficients as high as 3000 (hr)^{-1} based on the water phase concentrations. In countercurrent flow, since flooding puts a limit on maximum velocity, the mass transfer coefficients were limited to 100 (hr)^{-1} .

In cocurrent flow even higher coefficients could probably be attained with higher flow rates. Nearly equilibrium compositions were obtained in only a few inches

of packing. The mean mass transfer coefficient increased with the flow rates of both phases. They correlated their results by the following equation:

$$k_W = 0.00032 L^{1/2} V \quad (1)$$

where k_W = mean mass transfer coefficient for water in isobutanol-rich phase, $(\text{hr})^{-1}$

L = flow rate of continuous (water-rich) phase, $\text{lb.mole}/(\text{hr})(\text{sq.ft.column cross section})$

V = flow rate of dispersed (isobutanol-rich) phase, $\text{lb.mole}/(\text{hr})(\text{sq.ft.column cross section})$

1.2 Gas-Liquid Systems

Wen and co-workers(18) studied the dehumidification of air by calcium chloride solutions in packed beds. The liquid rate, $\text{lb}/(\text{hr})(\text{sq.ft.})$, was 40 - 100 times greater than the gas rate and the change in water concentration in the calcium chloride solution was negligible. Hence, the concentration versus bed height relationship in both the countercurrent and cocurrent flow patterns are quite similar. The mass transfer coefficient increased with the increasing gas rate but at constant gas rate it decreased with the increasing liquid rates. This decrease in transfer coefficient was probably due to the channeling which occurred at high liquid rates. Mass transfer coefficients up to $1300 \text{ lb-H}_2\text{O}/(\text{cu.ft.})(\text{hr})(\text{lb-H}_2\text{O}/\text{lb-air})$ were obtained.

Dodds and co-workers(3) studied the absorption of carbon dioxide in caustic soda solutions in packed beds. In this system the equilibrium partial pressure of carbon dioxide over caustic soda solutions is negligible and hence the driving force is similar to that in countercurrent flow. Higher transfer coefficients, up to 13 lb.moles/(hr)(cu.ft.)(atm.), were obtained by using flow rates greater than the flooding rates in countercurrent flow. The mass transfer coefficient K_a increased with increasing flow rates of gas and liquid.

Finzi and co-workers(6) studied the isothermal exchange of H₂O between liquid water and steam at atmospheric pressure in wetted wall columns. In the tube water occupied an annular ring and was raised by the gas friction. The mass transfer coefficient increased with the increasing ratio of molar liquid and gas flow rates. They report that throughput for columns designed using vertical upward annular cocurrent flow were 35 - 40 times greater than for the conventional countercurrent distillation units.

Tang and Himmelblau(15) studied the interphase mass transfer between carbon dioxide gas and water. The physical model used was the laminar cocurrent flow of gas and liquid in a rectangular channel of infinite width. The concentration distribution in the liquid phase was given by the following equation:

$$u \frac{\partial c}{\partial z} = D \frac{\partial^2 c}{\partial y^2} \quad (2)$$

where u = velocity of liquid in z direction
(direction of flow), cm/sec
 c = concentration of carbon dioxide in water
mg/cc
 y = distance from the gas liquid interface

This equation was solved with an assumed velocity distribution and proper boundary conditions for the diffusion coefficient D by three methods - the eigensolution method, the boundary layer theory and penetration theory. Results from the eigensolution agreed within 5 percent with the data of other investigators in the temperature range of 20° to 30°C. Calculated values of diffusion coefficients from the penetration theory checked well with those from the eigensolution method, but the values computed from the integrated boundary layer equation were about 10 percent higher.

Scott and Hayduk(13) studied the rate of gas absorption in cocurrently flowing gas-liquid systems in horizontal pipes. The variables studied were the superficial velocities of gas V_G and liquid V_L , liquid phase diffusivity D and viscosity μ , gas-liquid interfacial tension ∇ , gas density ρ and tube diameter d . The gas density had no effect on absorption measurements. At low liquid rates, the data showed a maximum in the number of transfer units with increasing gas velocity. This was not observed at high liquid rates and was due to the change in interfacial area. The number of

transfer units per unit length of absorber decreased nearly directly with increasing liquid flow rates. The correlation of data was presented by a logarithmic plot of N.T.U. per $(D^{0.5} \mu^{0.1} d^{2.0} V^{0.5})$ as ordinate against $V_G/(V_G + V_L)$ as abscissa.

2. Cocurrent Flow in Packed Beds

The recent papers describing the flow mechanism, pressure drop and phase holdup are reviewed in this section.

2.1 Liquid-Liquid Flow

Rigg and Churchill(11) studied the phase holdup, pressure drop and drop diameter in two phase flow. The experimental results indicated that steady state phase holdup can be approximated by assuming no slip velocity between the phases. An estimate of the phase holdup can be obtained by an equation of the form

$$H = \frac{U_o}{(U_o + U_w)^a} \quad (3)$$

where H = liquid holdup

U_o = organic phase superficial phase velocity

U_w = water phase superficial phase velocity

and a is an arbitrary constant evaluated for each system. A mixture of immiscible fluids flowing through a packed bed cannot be treated as a pseudo single phase with effective properties.

The observed data for pressure drop were correlated in terms of the parameter P_{ratio} , where P_{ratio} is the ratio of frictional pressure drop gradient to the pressure drop predicted by the assumption of single phase flow. The Sauter mean droplet diameter

$$d_{32} = \frac{\sum_{j=1}^n d_j^3}{\sum_{j=1}^n d_j^2} \quad (4)$$

is directly proportional to the packing diameter and can be correlated with velocity, viscosity and interfacial tension.

2.2 Gas-Liquid Flow

Scott(12) has given an excellent summary of the recent work on the properties of cocurrent gas-liquid flow. He has pointed out that the pressure drop and liquid hold-up for a two-phase system are influenced by the flow pattern.

Wen and co-workers(17) studied the pressure drop through packed beds for dehydration of air by calcium chloride solutions. They recognized three flow regions. The first or channeled region exists at low gas rates. At higher gas rates region II exists and in this region the flow mechanism can be assumed to be a thoroughly dispersed liquid phase throughout the continuous gas phase. Region III exists at high liquid rates and the liquid phase can be assumed to be the continuous phase with the gas phase completely dispersed throughout the liquid.

In Region I pressure drop is proportional to G^n , where G is the gas flow rate and $n = 2$ at low liquid rates and is less than one at high liquid flow rates. In Region II pressure drop varies approximately linearly with G . In Region III the power of G seems to be less than unity with possible variation. The pressure drop also depends upon the physical properties of fluids, packing size and configuration, phase distribution and flow pattern of the phases.

Weekman and Myers(16) also recognized the existence of three regimes in air-water system. They observed that similar relations correlate the pressure drop results for two phase flow in both pipes and packed beds.

Dodds and co-workers(4) studied the pressure drop and liquid holdup in cocurrent gas absorption in a packed tower and compared their results with countercurrent gas absorption. They concluded that the two flow systems are similar at lower flow rates. In each case logarithmic plots of pressure drop versus gas rate would give similar parallel lines per liquid parameter and hence the same exponent to be attached to gas rate. With countercurrent flow this exponent sharply increases at the flooding and loading point. Conversely in cocurrent flow this exponent may be decreased at higher gas rates, $\text{lb}/(\text{hr})(\text{sq.ft.})$.

In countercurrent tower operation, below the flooding point, liquid holdup is not affected by the gas rate. In cocurrent flow higher gas rates flatten the holdup to liquid-rate

relationship and the relative increase of holdup lessens at higher liquid rates.

3. Mass Transfer Studies for Benzene-Acetic Acid-Water System

The transfer of acetic acid between benzene and water has been studied in packed beds, wetted wall columns and spray columns. This section presents the nature and magnitude of the mass transfer coefficients obtained.

Sherwood and co-workers(14) studied the mass transfer of acetic acid from a water-rich phase to a benzene-rich phase in spray and packed columns. The mass transfer coefficient K_a was calculated from the equation

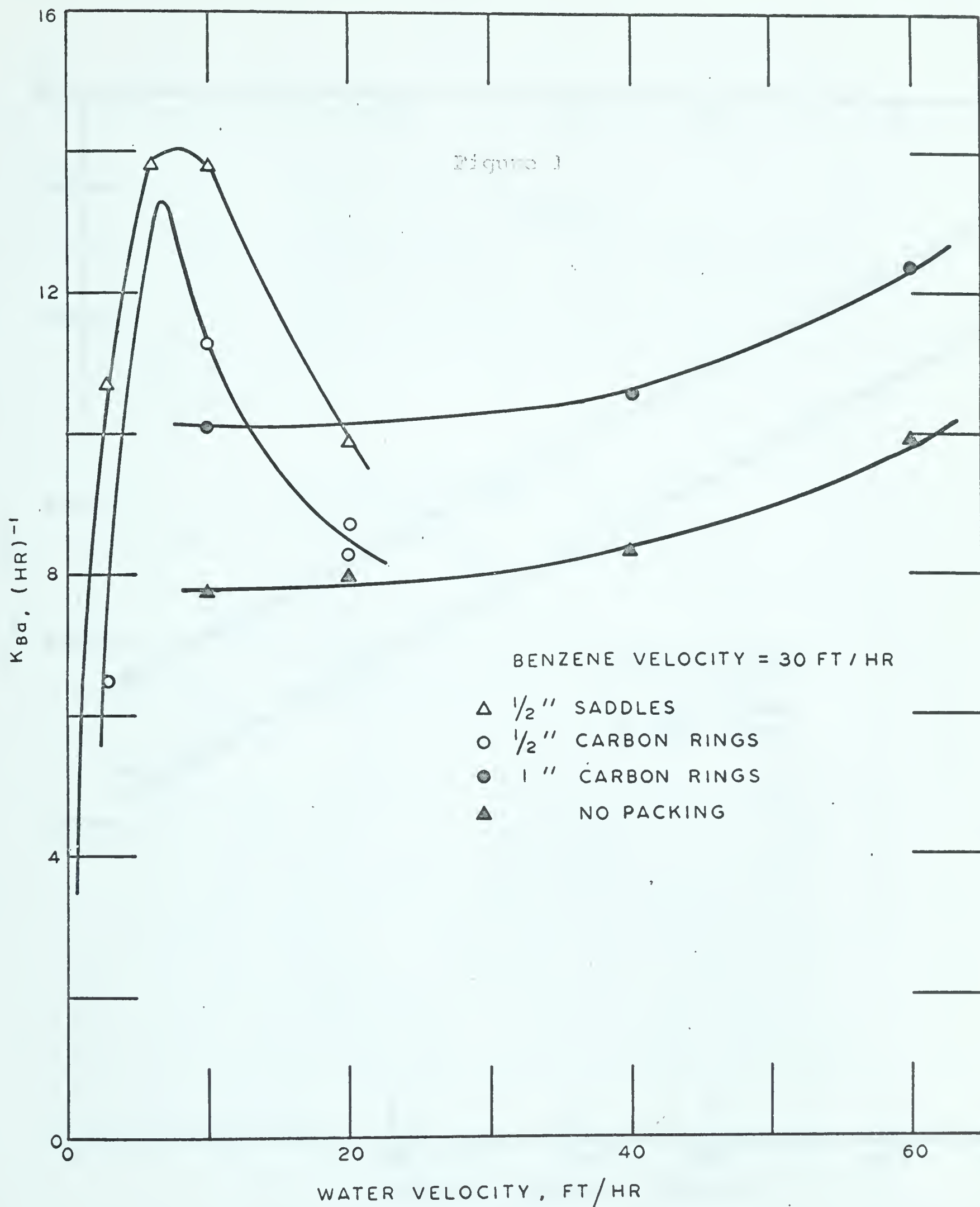
$$K_a = \frac{L (c_2 - c_1)}{A \Delta c_{1m}} \quad (5)$$

where c_2 and c_1 are the acid concentrations at exit and inlet respectively. The logarithmic mean driving force, Δc_{1m} is based on c_1 and c_2 and the equilibrium concentrations in benzene corresponding to the observed concentration in the aqueous phase at the bottom and top of the column.

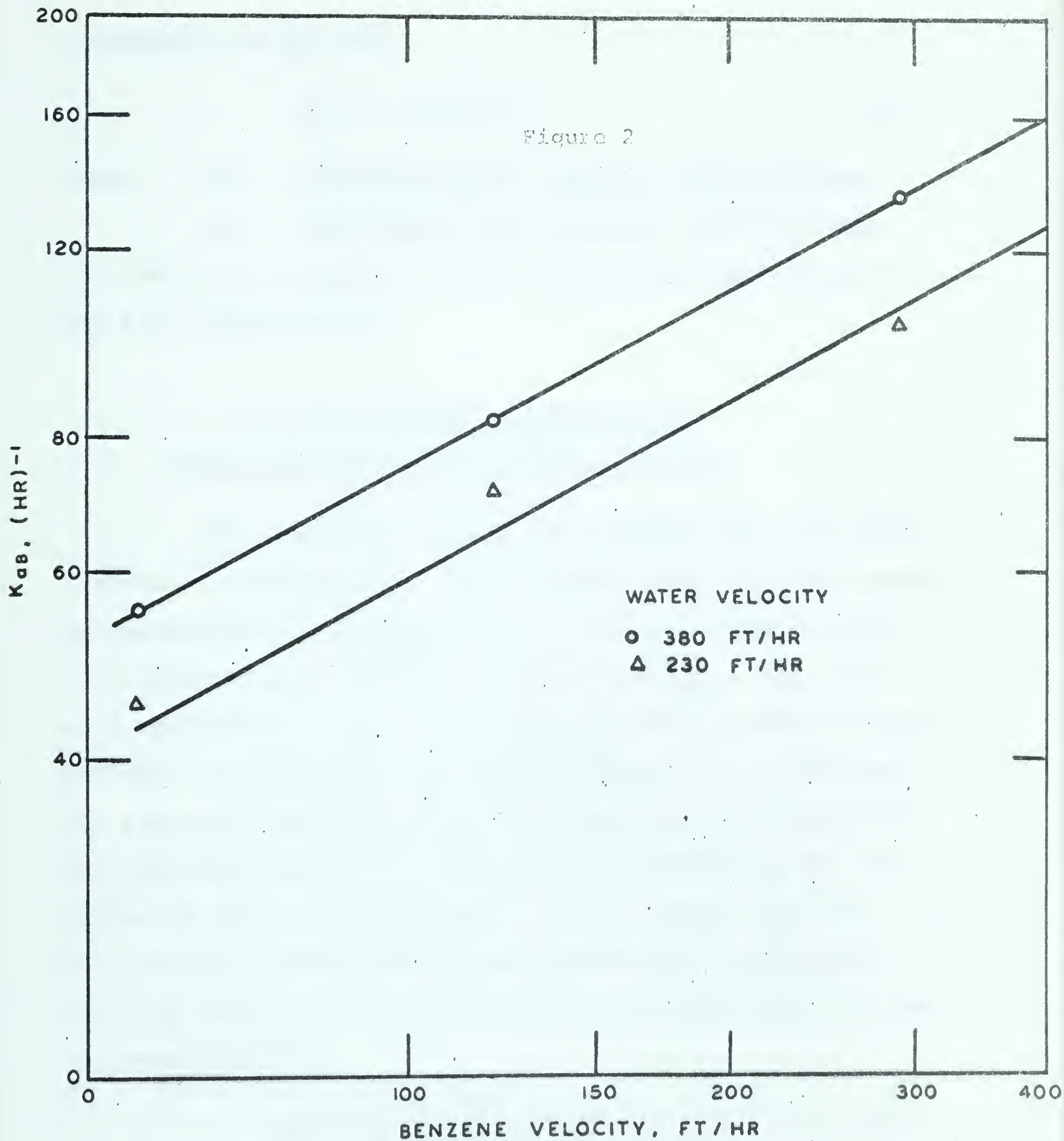
In spray columns mass transfer coefficients up to 1.6 (hr)^{-1} were obtained. The value of K_a increased with drop diameter because of the increased turbulence inside a large drop. The drop size in the packed bed, and hence the

value of K_a , depends more on the packing than on the size of the drops entering the packing. They concluded that variations in interfacial area a are more important than the effect of other variables. Their results for packed columns are shown in Figure 1. With increasing flow rates K_a passes through a maximum until coalescence offsets the advantage of large hold-up at high flow rates. The 1-inch carbon rings served only to increase the holdup somewhat as compared with the operation without packing.

Comings and Briggs(1) studied the mass transfer of acetic acid from a benzene-rich phase to a water-rich phase in packed and wetted wall columns. In packed columns no indication of maximum K_a at velocities below the flooding point was observed. As compared to Sherwood and co-workers(14) they obtained higher mass transfer coefficients, up to 31 (hr)^{-1} , by using shorter packed columns. Such results may be due to the large proportion of extraction taking place at the nozzle inlet or to a greater tendency for the longer columns to channel at low velocities. Dispersed phase velocity had a greater effect on the overall mass transfer coefficient. Their results for wetted wall columns are shown in Figure 2. In wetted wall columns values of K_a as high as 134 (hr)^{-1} were obtained by using flow rates which were about 30 times greater than for the packed columns. They proposed that overall coefficients for small packed columns may be represented by



Extraction of Acetic Acid in Countercurrent Flow
in a 3.55 inches Diameter and 66 inches High Packed Column



Extraction of Acetic Acid in Countercurrent Flow
in a 0.85 inch Diameter and 46.5 inches High Wetted Wall Column

an equation of the form

$$K_a = c V_D^r V_C^s \quad (6)$$

where V_D = dispersed phase velocity, $(\text{ft}^3)/(\text{ft}^2)(\text{hr})$

V_C = continuous phase velocity, $(\text{ft}^3)/(\text{ft}^2)(\text{hr})$

c , r and s are constants. Their values for r and s were 0.5 and 0.25, respectively.

4. Mass Transfer Studies in the Ethyl Acetate-Acetic Acid-Water System

The transfer of acetic acid between water and ethyl acetate in countercurrent flow in packed beds has been studied by Eaglesfield and co-workers(5). A 5 percent aqueous acetic acid solution was extracted with ethyl acetate. The acetic acid concentration was determined by titrating against sodium hydroxide solution and using phenolphthalein as an indicator. The flow rates were calculated by collecting the exit extract and raffinate phases for a known time and measuring the densities of inlet and exit phases. It was assumed that the mass transfer coefficient K_a was constant for a given flow ratio of solvent to water and its value was calculated by the following formula.

$$K_a = \frac{N/\theta}{V (\log \text{ mean } \Delta c)} \quad (7)$$

where N/θ = gm mole of acetic acid transferred per hour
 V = volume of packed space
 Δc = effective concentration difference in gm mole per liter at top and bottom of the column, calculated from the relation $\Delta c = c_e - c_e^*$
where c_e = concentration of acetic acid in extract phase
 c_e^* = equilibrium concentration of acetic acid corresponding to the raffinate composition

All extractions were carried out at about 70 percent of the flooding velocities and the maximum value of K_a based on the concentrations in water-rich phase was 5.6 (hr)^{-1} . The K_a values were higher when the aqueous phase was dispersed.

The authors(5) also studied the economics of the recovery of acetic acid from dilute aqueous solutions obtained from various industrial processes. On the basis of steam consumption calculations they concluded

"Below about 16 percent by weight of acetic acid, extraction with ethyl acetate is the most economic method of recovery. From 16 to about 25 percent by weight of acetic acid, extraction with a mixture of ethyl acetate and benzene is most economic. The optimum benzene concentration is approximately $1.67(x - 16)$ percent by volume, where x is the weight percent of acetic acid in the feed. From 25 to 35 percent by weight of acetic acid, the most economic recovery process is solvent extraction with isopropyl acetate. Above 35 percent it is more economic to recover the acetic acid by

an entrainment distillation process, preferably with the use of an Othmer entrainer such as 80 percent isoamyl acetate per 20 percent isoamyl alcohol."

For extracting acetic acid, benzene has a greater selectivity (its ability to extract one component of a solution in preference to another) and ethyl acetate has a larger distribution coefficient

$$m = \frac{c_o}{c_w} \quad (8)$$

where c_o and c_w are the concentrations of acetic acid in organic-rich phase and water-rich phase, respectively. At high concentrations of acetic acid the low selectivity of ethyl acetate is improved by the addition of benzene.

5. Solubility and Tie Line Data

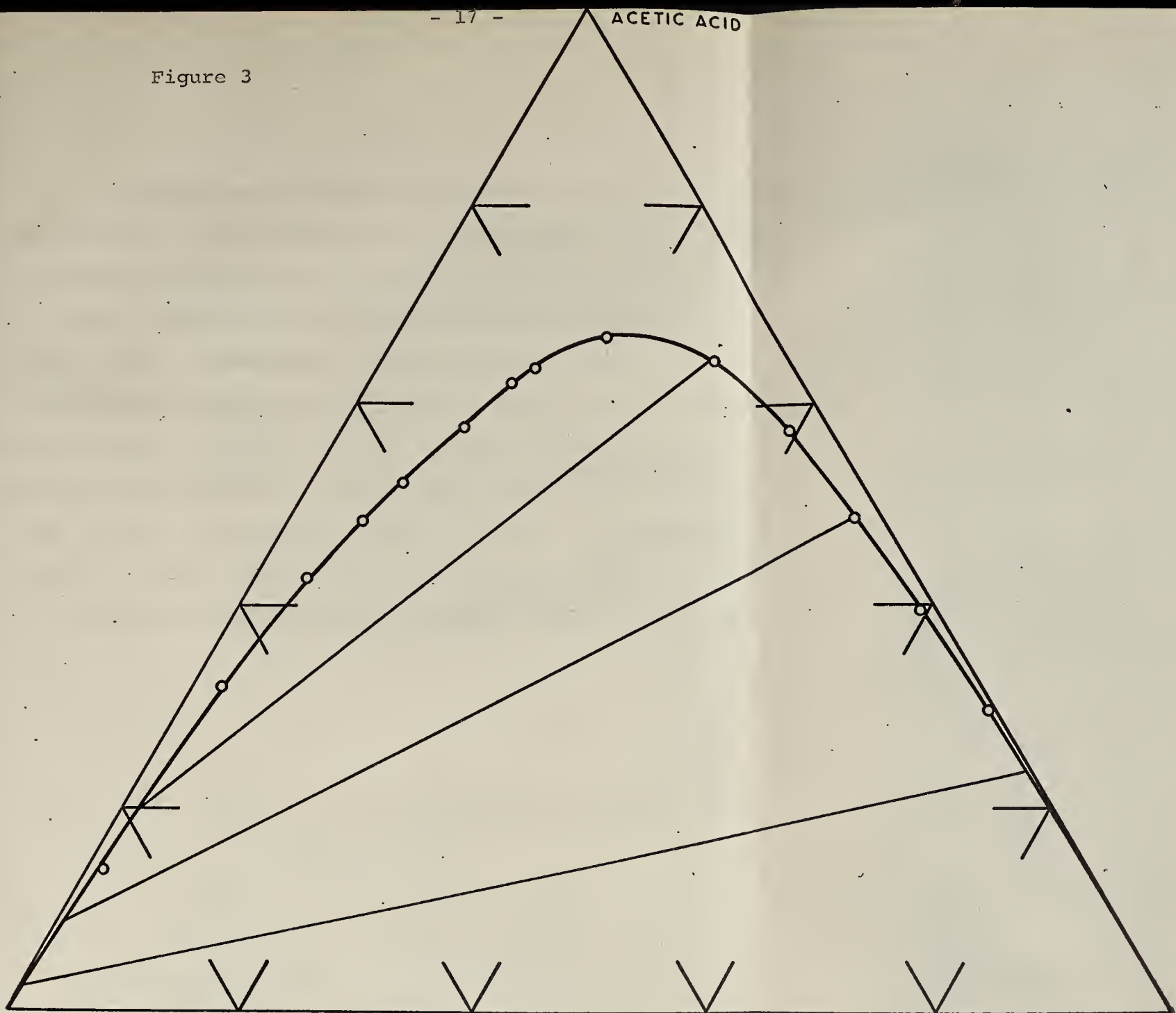
Garner and co-workers(7) studied the solubility and tie line data for the water-acetic acid-benzene system between 30° and 60°C. The solubility curve and tie lines at 30°C are shown in Figure 3. Their results indicated that solubility changes only slightly with temperature. The distribution coefficient shows only a small variation with temperature when the concentration of acetic acid in the water layer is less than 50 percent by weight. The tie line data was satisfactorily correlated by Bachman and Othmer - Tobis plots.

Figure 3

BENZENE

WATER

Solubility and Tie line Data for the Benzene-Acetic Acid-Water System at 30°C

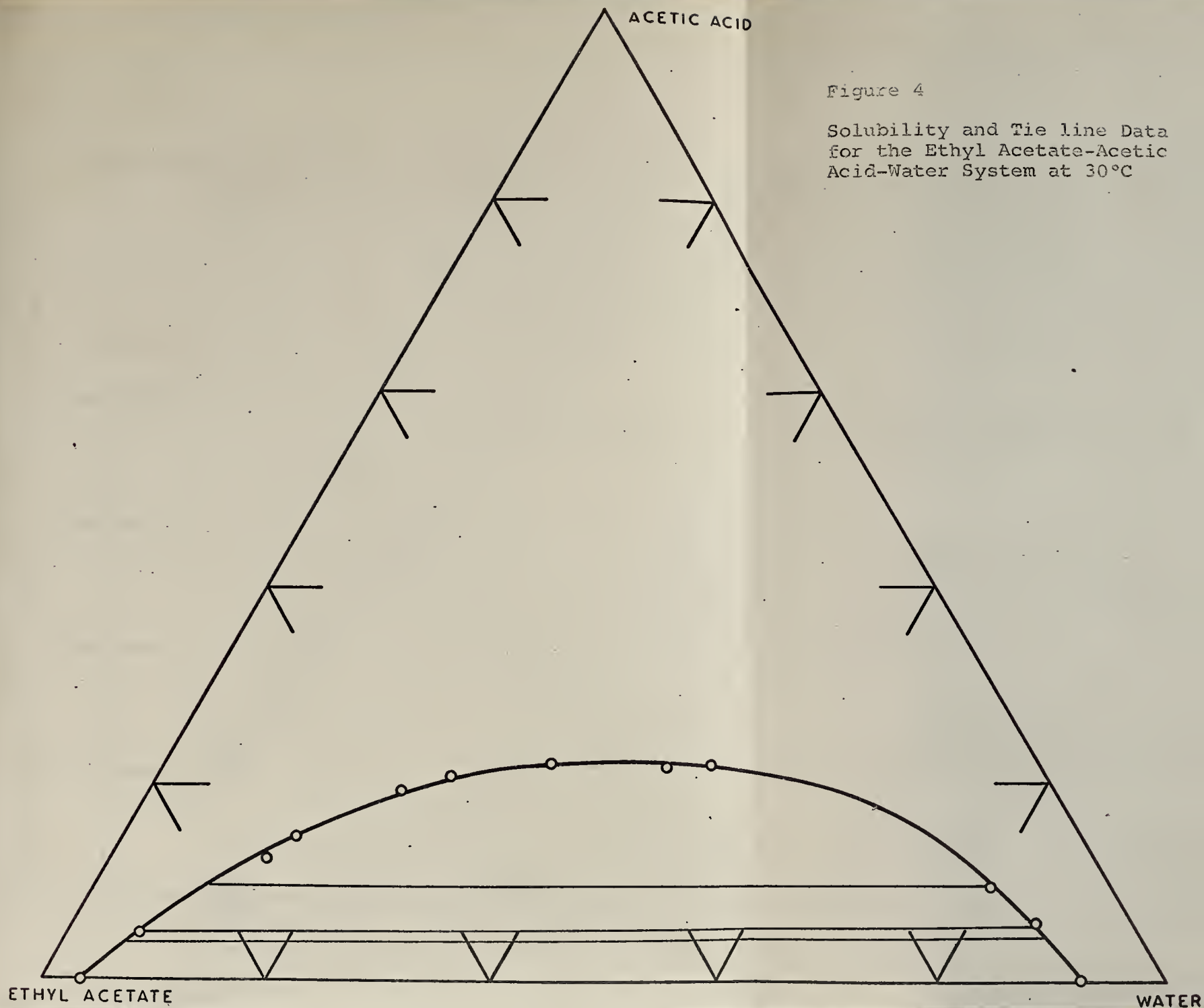


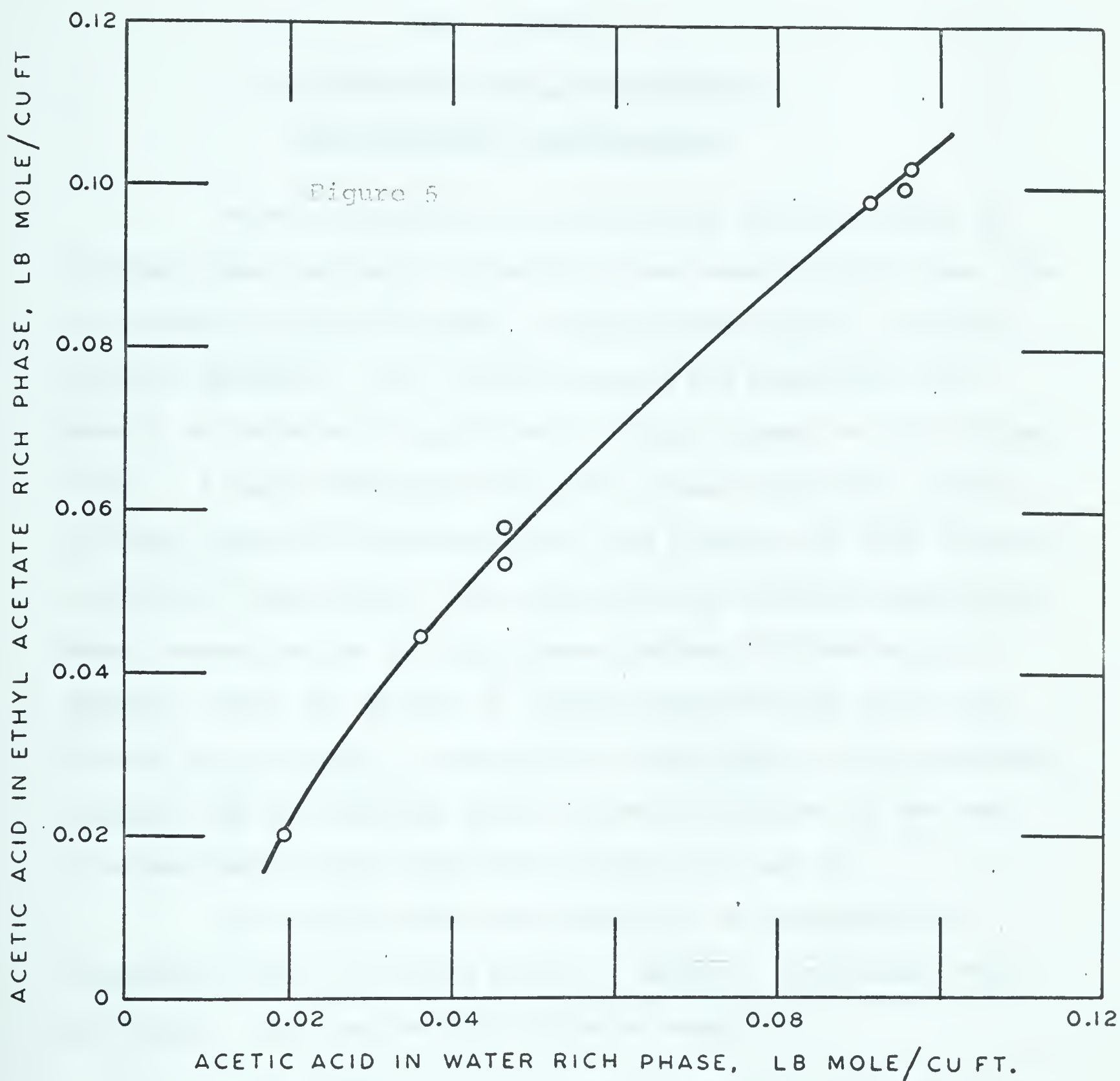
Garner and Ellis(8) have studied the solubility and tie line data for the water-acetic acid-ethyl acetate system. The solubility curve and tie lines at 30°C are shown in Figure 4. Their results indicated that solubility changes only slightly with temperature. The equilibrium data is almost independent of temperature changes in the range of 30° to 60°C and is shown in Figure 5. Their results agreed well with those of other workers. The selectivity of acetic acid for ethyl acetate reverses at a certain stage and then becomes selective towards water and thus this system is a solutrope at 9.5 percent by weight of acetic acid in the water layer.

ACETIC ACID

Figure 4

Solubility and Tie line Data
for the Ethyl Acetate-Acetic
Acid-Water System at 30°C





Equilibrium Data for the Ethyl Acetate-Acetic Acid Water System

III. THEORY

1. Definition and Calculation of Mass Transfer Coefficients

When a material is transferred from one phase to another phase across an interface that separates the two, the resistance to mass transfer in each phase causes a concentration gradient. For a given system and conditions the amount of material transferred is proportional to the driving force. A more efficient operation results from the greater driving force of countercurrent flow (Figure 6a) than that of cocurrent flow (Figure 6b) with the same terminal conditions when concentration in both phases varies with bed height. However, when the change in liquid concentration with bed height in one phase is constant or negligible, this advantage ceases, and the driving force for mass transfer in the two systems tends to be equivalent (Figure 6c and d).

At steady state the diffusion of transferring component from one liquid phase to another, immiscible with the first, is given by the following equation:

$$dN = k_R (c_R - c_{Ri}) dS = k_E (c_{Ei} - c_E) dS \quad (9)$$

where N = rate of mass transfer, lb.mole/(hr)(sq.ft.)
 k = individual mass transfer coefficient,
lb.mole/(hr)(sq.ft.)(Δc)
 c_R = concentration of transferring component in
raffinate phase, lb.mole/(cu.ft.)

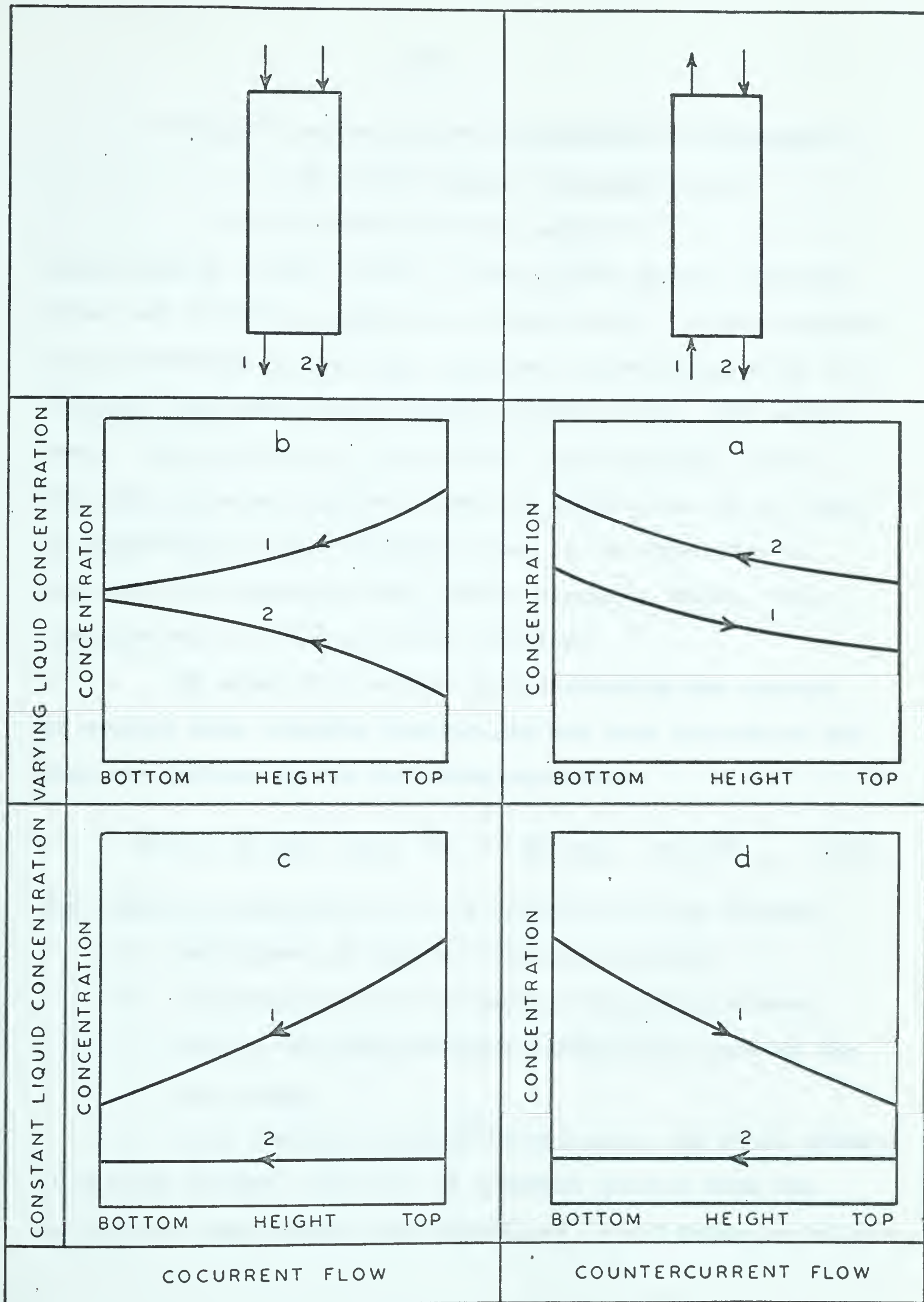


Figure 6

Phase Concentrations versus Bed Height Relationship in Cocurrent and Countercurrent Flow Processes

c_E = concentration of transferring component
in extract phase , lb.mole/(cu.ft.)

S = interfacial area, sq.ft.

Subscripts E, R and i refer to the extract phase, raffinate phase and interface conditions respectively. In this equation it is necessary to know the interface concentrations for calculating individual mass transfer coefficients. The experimental determination of interfacial concentrations is not possible since any ordinary sampling device will be so large in comparison to film thickness that it is impossible to approach the interface sufficiently closely. Hence, this equation has a little practical utility.

In order to overcome this difficulty the concept of overall mass transfer coefficient has been introduced and they are defined by the following equations:

$$dN = K_R (c_R - c_{Re}) dS = K_E (c_{Ee} - c_E) dS \quad (10)$$

The implicit assumptions in this equation were as follows:

1. Resistance of the two films are additive
2. Concentrations of the contact interface correspond to the equilibrium concentrations between the two phases.

It is usually possible to calculate the total surface of the dry packing, but this is somewhat greater than the interfacial area because the circulated liquid tends to collect

at the points of contact of the lumps of packing and all the surface may not be wet. For this reason it is convenient to introduce a new variable, a , which represents the interfacial area per unit of volume and

$$dS = a dz \quad (11)$$

where z is the height of the packed column. Since both a and K depend on the nature of the packing and on the liquid flow rates, they may be combined as Ka , which represents the overall capacity coefficient, on a volume basis, for any particular packing. Equation 10 then becomes

$$d(v_E c_E) = Ka_E (c_{Ee} - c_E) dz \quad (12)$$

where Ka = overall mass transfer coefficient,
lb.mole/(hr)(cu.ft.)(Δc)

v = superficial phase velocity, cu.ft./(hr)(sq.ft.
of the cross sectional area of packed column).

If v_E does not vary in the packed column appreciably, then it can be treated as a constant. When it is possible to assume that the equilibrium curve is linear over the range in which it is to be used then Ka_E does not depend on the concentration c_E . Equation 12 then becomes

$$\int_{z_2}^{z_1} dz = \frac{v_E}{Ka_E} \int_{c_{E2}}^{c_{E1}} \frac{dc_E}{(c_{Ee} - c_E)} \quad (13)$$

If the operating line is also linear over the range in which it is to be used then equation 13 can be integrated to give

$$(z_1 - z_2) = \frac{v_E}{K_{aE}} \frac{(c_{E1} - c_{E2})}{(\Delta c_E)_{lm}} \quad (14)$$

where $(z_1 - z_2)$ = height of the packed column, and

$$(\Delta c_E)_{lm} = \frac{(c_{Ee} - c_E)_1 - (c_{Ee} - c_E)_2}{\ln \left[(c_{Ee} - c_E)_1 / (c_{Ee} - c_E)_2 \right]} \quad (15)$$

subscripts 1 and 2 refer to the properties at the top and bottom of the packed column. On rearranging, equation 14 becomes

$$K_{aE} = \frac{v_E}{(z_1 - z_2)} \frac{c_{E1} - c_{E2}}{(\Delta c)_{lm}} \quad (16)$$

This equation has been used in this work to calculate the mass transfer coefficients.

2. Correlations for Mass Transfer Coefficients

In film theory the resistance to mass transfer is described by a combination of laminar film and turbulent zone, including the intervening buffer zone. Thus, the total resistance to mass transfer is made of two parts: that residing in the laminar film and that represented by the buffer zone and turbulent region. The diffusional resistance of the

laminar and turbulent films can be related to the Reynolds number. The molecular diffusivity D will be necessary to describe the rate of diffusion across the laminar film but it is of little importance in considering the turbulent region. The mass transfer coefficient, therefore, can be expected to depend upon the diffusivity, D , and the variables making up the Reynolds number, the average fluid velocity u_{av} , the density ρ , viscosity μ and some characteristic dimension of the apparatus d . Thus,

$$k = \phi (D, u_{av}, d, \mu, \rho) \quad (17)$$

or, if arranged in terms of dimensionless groups,

$$\frac{kd}{D} = \phi' \left(\frac{d u_{av} \rho}{\mu}, \frac{\mu}{\rho D} \right) \quad (18)$$

$$= \phi' (Re, Sc) \quad (19)$$

On the assumption that the degree of turbulence in each phase is affected by the rate of flow of both phases, since the liquids are in direct contact, equation 19 becomes

$$\frac{k_R d}{D_R} = \psi (Re_R, Re_E, Sc_R) \quad (20)$$

At constant temperature the physical properties of fluids and hence Sc are constant and equation 20 becomes

$$\frac{k_R d}{D_R} = \psi' (Re_R, Re_E) \quad (21)$$

But the Reynolds number cannot always be clearly defined in packed columns, and furthermore it is not usually so clear an indication of the degree of turbulence as in simpler fluid systems. Hence, for a given apparatus and liquid system, whose fluid properties vary little with operating conditions, equation 21 may be written as follows:

$$k_R = \psi''(u_R, u_E) \quad (22)$$

According to two-film theory the relative importance of the two additive resistance depends on the solubility coefficient as well as on the nature of two films. It would therefore be expected that the overall mass transfer coefficient will depend on the solubility and all other factors which affect the individual mass transfer coefficient.

In addition to the above variables the overall mass transfer coefficient in packed beds may also depend on the following variables.

1. Packed bed height
2. Entrance or mixing arrangement
3. Packing shape and size
4. Column size
5. Direction of extraction, etc.

There is no satisfactory general correlation for the overall mass transfer coefficient.

IV. EXPERIMENTAL PROGRAM

This section describes in detail the experimental equipment and operating procedures used to study some of the variables affecting the mass transfer rate. It also describes the manner of selection and range of variables studied.

1. Experimental Apparatus

The principal part of the apparatus for the present study was the packed column. The column was a 5/8-inch nominal diameter glass pipe cut to the desired lengths. The diameter of the column was chosen to permit high mass velocities without the use of excessive amounts of solvents. No attempt was made to recover the spent solvents.

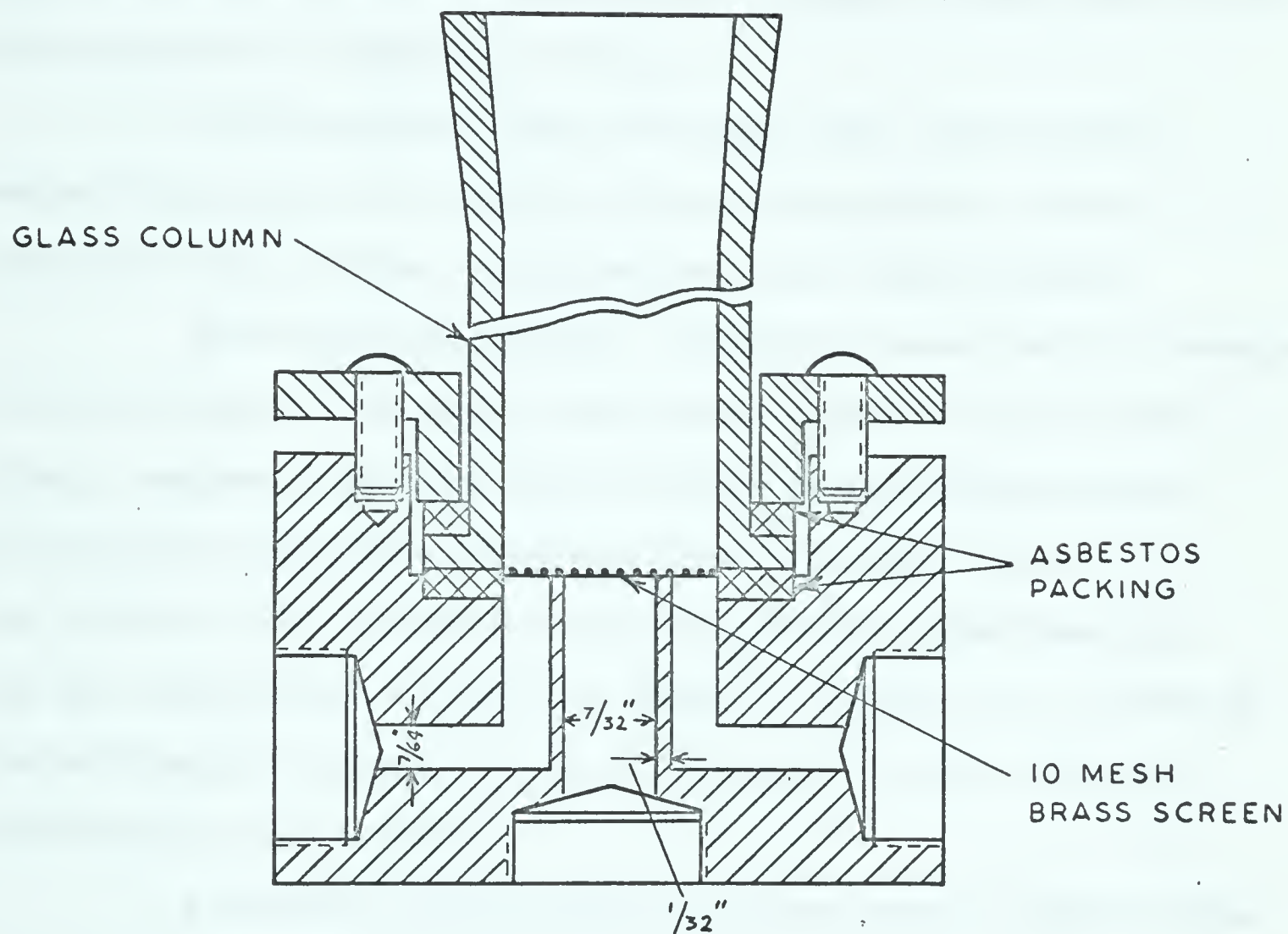
The packing material was 3 mm diameter solid glass beads. These beads were supplied by the Fisher Scientific Company and were graded to get a uniform packing. The diameter of glass beads used for packing was between 3.327 and 2.794 mm. The glass beads were cleaned by chromic acid and thoroughly washed with water. The beads were held in place by a 10-mesh brass screen at the top and bottom of the column.

The column inlet flange was similar to that used by Leacock and Churchill(10). They introduced both phases into the packed column through two adjacent channels in the inlet flange. Their design was modified for this work by

introducing the organic-rich phase through a central channel and the water-rich phase through two side channels. This modification was done to hopefully minimize the entrance effects. A detailed drawing of the inlet flange and glass column is shown in Figure 7. The inlet flange was made of aluminum. A 1/4-inch stainless steel valve was used to control the distribution of the water-rich phase in the two channels. This particular design of inlet flange eliminated any mass transfer before the liquids entered into the packed column.

A round-bottom glass flask, equipped with a short side arm and a flange at the equator so that it could be connected to the packed column, was used as a settler. It was connected to the packed column by 5/8-inch cast iron backing flanges. Asbestos gaskets with fluon gasket sheaths were used. It also had two openings at the top and bottom for the removal of the two phases. Samples were taken at these two outlets. In the bottom discharge line a stopcock was used to adjust the level of the interface between the two phases in the settler. A thermometer, range -30 to 120°F, was used to determine the temperature at the middle of the settler and this temperature was then taken as being equal to the temperature in the packed column. The settler was connected to the drain pipe by saran tubes. Three settlers of 200, 300 and 500 ml capacity were prepared so that an appropriate size could be used at different flow rates.

Figure 7



COLUMN INLET FLANGE DETAILS

A 1/4 H.P. Duplex Milton Roy Controlled Volume Diaphragm pump, Model R-231 of 100 - 400 cc/min. capacity was used to pump the two phases. The discharge rate of the pump was controlled by adjusting the volume of hydraulic fluid which bypassed a diaphragm cavity.

The pulsations from the pump were successfully damped by passing the liquids through dampeners. These dampeners were 1-liter, round-bottom pyrex glass flasks.

Two Fisher and Porter rotameters were used to measure the flow rates. The water-rich phase rotameter was of 540 cc/min. maximum capacity and was made from stainless steel. The organic-rich phase rotameter had a maximum capacity of 550 cc/min. The rotameters were calibrated experimentally and the calibration curves are shown in Figures 17, 18 and 19. The calibration curves agreed substantially with those provided by the manufacturer.

A constant temperature bath was used to control the temperature of liquids entering in the packed column. This constant temperature bath was a 12-inch x 12-inch cylindrical heavy-walled pyrex brand glass jar. The bimetallic thermostat, range -100 to 350°F, and electronic relay were purchased from the American Instrument Company Inc. A 300-watt heater was used. The stirrer had a 1/30 H.P. Edison Universal motor. A thermometer, range -20 to 120°F, was used to indicate the bath temperature. A pilot light was provided

which indicated the operation of intermittent heater. The temperature of the bath could be controlled within $\pm 0.5^\circ\text{F}$.

The water-rich phase and ethyl acetate-rich phase reservoirs were twelve-gallon and five-gallon pyrex glass bottles, respectively. The benzene phase reservoir was a five-gallon mild steel drum and was provided with a level gauge glass. A schematic diagram of the experimental apparatus is shown in Figure 8.

The size of the inlet lines to the pump was 3/8-inch O.D. and all other lines were 1/4-inch O.D. tubes. A 1/4-inch bronze strainer was used on the organic-rich phase pump inlet line. Stainless steel and copper tubes were used for the water-rich and organic-rich phases, respectively. All valves with the exception of a 3/8-inch Jenkins bronze valve in the organic phase inlet line to the pump were Hoke stainless steel valves of the 3,000 psi series. All connections were made by compression fittings. The apparatus was mounted on a dexion framework.

2. Experimental Determination of Equilibrium

Date for Benzene-Acetic Acid-Water System

The concentration of acetic acid in the benzene-rich phase in this work varied from 0.2 to 0.8 percent by weight and extrapolation of the data of Garner and co-workers(7) was considered unreliable, therefore, equilibrium data were deter-

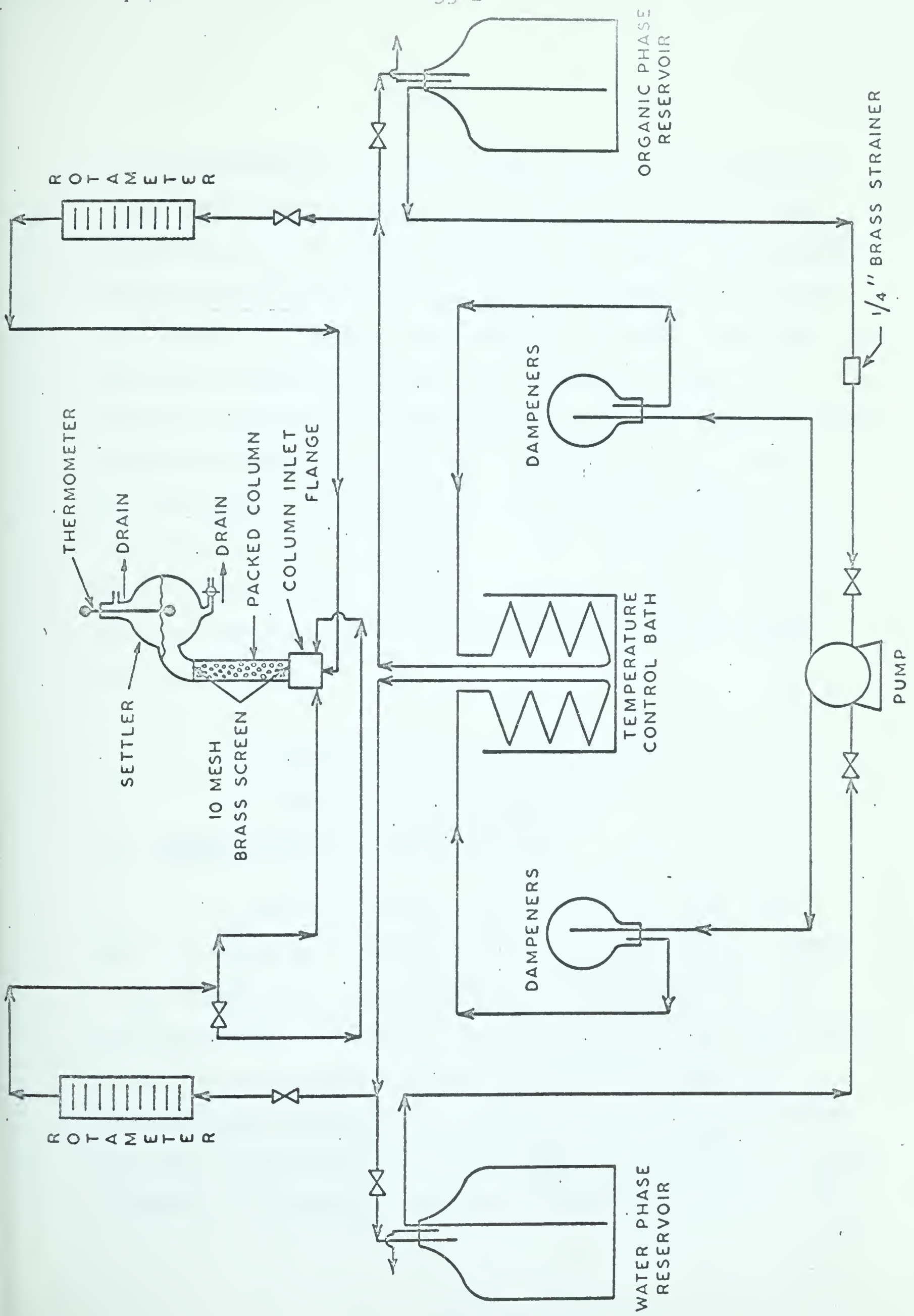


Figure 8 SCHEMATIC DIAGRAM OF EXPERIMENTAL APPARATUS

mined experimentally for this range. Acetic acid solutions of different concentrations were prepared by adding a calculated quantity of acetic anhydride into water. Equilibrium mixtures were prepared in test tubes by mixing 50 cc of benzene and 50 cc of acetic acid solution in water. The test tubes were stoppered and were then allowed to stand in a constant temperature bath maintained at $86 \pm 0.5^{\circ}\text{F}$ for 20 hours. Samples from the water-rich and benzene-rich phases were titrated against standard sodium hydroxide solutions using phenolphthalein as an indicator. After allowing these test tubes to stand in the constant temperature bath for a longer time, no change in concentration of acetic acid in either phase was detected.

3. Experimental Determination of Mass Transfer Coefficient

3.1 Benzene-Acetic Acid-Water System

A layer of benzene was maintained at all times above the solution in the water-rich phase reservoir. After adding benzene the solution was stirred manually and then recirculated for two hours. It was then left overnight before making the actual runs. Thus, the water-rich phase was saturated with benzene at all times. As pointed out by Sherwood and co-workers(14) the equilibrium concentration of water in benzene is negligible and thus it was not necessary to mix

water in benzene.

The column, when dry, was packed with dry packing. No settling of packing was observed at the end of runs. The general procedure for making a run was as follows:

1. The pump was started and liquids were pumped through the dampeners, constant temperature bath, rotameters and the packed column.
2. The temperature in the packed column, as indicated by the temperature in the settler, was controlled at $86 \pm 2^{\circ}\text{F}$ by the constant temperature bath.
3. The level of the interface between the two phases in the settler was adjusted by using a stopcock to get the best separation.
4. Preliminary tests revealed that the settler outlet compositions attained steady state values after an operating period corresponding to one and a half volumetric displacements of the settler. For this work the phases were sampled after three displacement periods.
5. Flow rates were recorded. The outlet phases were sampled and analyzed.

3.2 Ethyl Acetate-Acetic Acid-Water System

The water-rich and ethyl acetate-rich phases were kept saturated with ethyl acetate and water, respectively by

using a similar procedure as used for the previous system. The solubility of water in ethyl acetate at 30°C is 3.5 percent by weight and thus it was necessary to saturate the ethyl acetate-rich phase.

The general procedure for making a run was exactly the same as used for the first system. In this system the steady state concentrations were attained after one displacement period and the samples were taken after two displacement periods.

4. Sampling and Analysis

Samples were collected in a 200-ml dry conical flask. The flask was then stoppered to prevent any change in concentration by evaporation. The organic-rich and water-rich phases were titrated within 8 and 20 hours, respectively.

All concentrations were determined volumetrically by titrating acetic acid against a standard solution of sodium hydroxide using phenolphthalein as an indicator. Duplicate titrations were performed for each sample.

The benzene-rich phase was analyzed by shaking 10 cc portions with 25 cc of distilled water and titrating the mixture with N/40 standard sodium hydroxide solution, shaking violently until the end point was reached. A faint pink color in the water layer was taken as the end point. It was found that the same end point was obtained when sufficient

alcohol was added to make the two phases miscible.

Ethyl acetate-rich solution was titrated against 1-N sodium hydroxide solution by a similar procedure as used for the benzene-rich solution.

The water-rich phase was titrated against 1-N standard sodium hydroxide solution. A faint pink color was taken as the end point.

Preliminary tests showed that the change in concentration of sodium hydroxide solution is negligible within 24 hours. The normality of standard sodium hydroxide solution was determined by titrating against two standard oxalic acid solutions and the results were accepted when they checked within ± 0.5 percent.

5. Properties of Materials

All chemicals used in this work were purchased from the Fisher Scientific Company. The acetic acid was prepared by adding a calculated quantity of acetic anhydride in distilled water. The acetic anhydride met the A.C.S. standards.

Purified benzene, boiling point 80 - 82°C, was used. Use of this benzene, within the experimental error, gave the same results as obtained by using the high purity benzene which conformed to the A.S.T.M. specifications.

Purified ethyl acetate was used which met the N.F. specifications.

V. EXPERIMENTAL RESULTS.

The experimental work was carried out to investigate the influence of phase velocities on the overall mass transfer coefficient for the systems benzene-acetic acid-water and ethyl acetate-acetic acid-water. For the system benzene-acetic acid-water, the effect of bed height on the overall mass transfer coefficient was also studied. Table I lists the number of runs and the range of variables studied. The temperature in the packed column for all observations was maintained at $86 \pm 2^\circ\text{F}$.

Overall mass transfer coefficients were calculated by using the following formula

$$K_{ao} = \frac{V_o}{z} \frac{(c_{o1} - c_{o2})}{(\Delta c_B)_{l.m}} \quad (23)$$

where K_{ao} = overall mass transfer coefficient based on organic-rich phase concentrations,
lb.mole/(hr)(cu.ft.)(Δc)
 z = packed bed height, ft.
 V = phase velocity, cu.ft./(hr)(sq.ft.)
 c_{o1} = concentration of organic-rich phase at the inlet, lb.mole/(cu.ft.)
 c_{o2} = concentration of organic-rich phase at the outlet, lb.mole/(cu.ft.)
 $(\Delta c_B)_{l.m}$ = logarithmic mean of terminal potentials, defined by equation 15.

TABLE I

Number of Runs and Range of Variables Studied

System	Variable Studied	V_O	V_W	z	No. of Runs
I	Effect of water rich phase velocity	77 - 440	130 - 486	5.9	31
I	Effect of benzene rich phase velocity	77 - 460	111 - 467	4.9	15
I	Effect of bed height	180 - 209	165 - 317	1.1 - 5.9	8
II	Effect of ethyl acetate-rich phase velocity	113 - 447	135 - 478	5.9	8
II	Effect of water-rich phase velocity	113 - 447	135 - 478	5.9	8

where System I = benzene-acetic acid-water

 System II = ethyl acetate-acetic acid-water

V_O = organic-rich phase velocity,
 cu.ft./ (hr) (sq.ft.)

V_W = water-rich phase velocity,
 cu.ft./ (hr) (sq.ft.)

z = bed height, inches

The implicit assumption in using the logarithmic mean terminal potential is that the operating and equilibrium curves are straight lines over the range of concentrations studied. Constant flow rates were used in the calculation of mass transfer coefficients.

1. Visual Observations

In addition to the numerical data presented later in this section, visual observations of flow in the packed column and settler were recorded.

The liquid mixtures in the packed column had a "milky" appearance and separated very rapidly after leaving the packing. Mass transfer in the outlet line and settler would therefore be expected to be negligible. This assumption was checked experimentally for the benzene-acetic acid-water system. During the course of experiments it was extremely difficult to determine which of the two phases was continuous. Since in all cases the water-rich phase preferentially wetted the glass packing it was thought to be continuous.

The benzene and ethyl acetate-rich phases in the settler were clear at all times but tiny droplets of this phase were observed in the water-rich phase. Groothuis and Zuiderweg(9) have suggested that this phenomena is a result of gradients in the interfacial tension caused by non-uniform mass transfer along the droplet interface. As proposed by

Davis and Rideal(2) some of the organic phase droplets in the water-rich phase may be present due to spontaneous emulsification. The spontaneous emulsification takes place by diffusion and stranding.

When the water-rich phase was allowed to stand, a thin film of benzene-rich phase was formed on the surface. In all runs the outlet compositions were close to the equilibrium value, therefore any change in the water phase concentration due to this thin film of benzene should be negligible. Moreover, only the equilibrium concentration in the benzene-rich phase corresponding to the water-rich phase concentration was used in mass transfer coefficient calculations. The equilibrium concentrations are not sensitive to a small change in the water-rich phase concentration. Thus, this thin film of organic phase on the water-rich phase should not have any appreciable effect on the value of mass transfer coefficients.

2. Benzene-Acetic Acid-Water System

2.1 General

In this study the concentration of acetic acid in the water-rich phase varied from 6.3 to 8.7 percent by weight and in the benzene-rich phase it was between 0.1 to 0.34 percent by weight. Figure 3 shows that in the above range of acetic acid concentration there is no significant mass transfer of water and benzene. Hence, acetic acid was considered as the

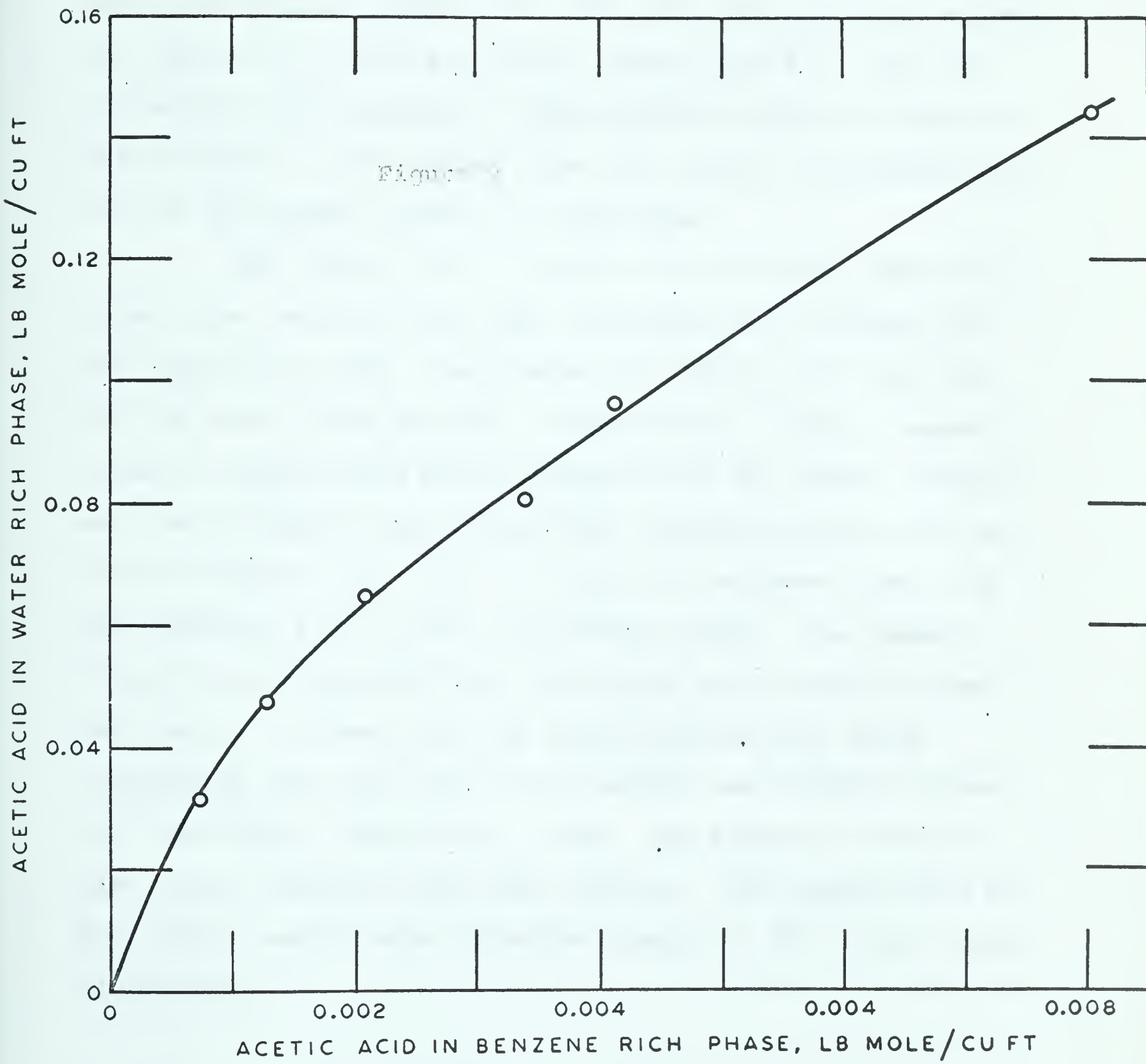
only transferring component. The change in flow rate was negligible due to the small amount of transferring acetic acid, therefore, inlet flow rates were used for calculations.

The equilibrium distribution of acetic acid between water and benzene favors the water-rich phase and therefore one would expect that major resistance to mass transfer is in the benzene phase. The equilibrium distribution varies with concentration and the magnitude of c_W/c_B is approximately equal to 30(7) where c_W and c_B are the concentrations of acetic acid in water-rich and benzene-rich phases, respectively. It was therefore expected that the benzene velocity would have a greater effect on the mass transfer coefficient. This was confirmed by the experimental results.

2.2 Equilibrium Data

The concentration of acetic acid in the water-rich phase during the course of experiments varied from 0.091 to 0.066 lb.mole/(cu.ft.). The equilibrium curve in Figure 9 shows that in the range of acetic acid concentrations studied the equilibrium data can be plotted as a straight line.

The experimentally determined equilibrium data is shown in Table IV and a sample calculation is given in Appendix C.



Equilibrium Data for the Benzene-Acetic Acid-Water System

2.3 Mass Transfer Outside of the Packed Column

Initially eight runs at different velocities were performed without packing in 2.532 and 4.935 inch high columns. The analysis of inlet and outlet phases failed to show any appreciable mass transfer. These results indicated that the mass transfer at the column inlet and outlet, as compared with that in the packed column, is negligible.

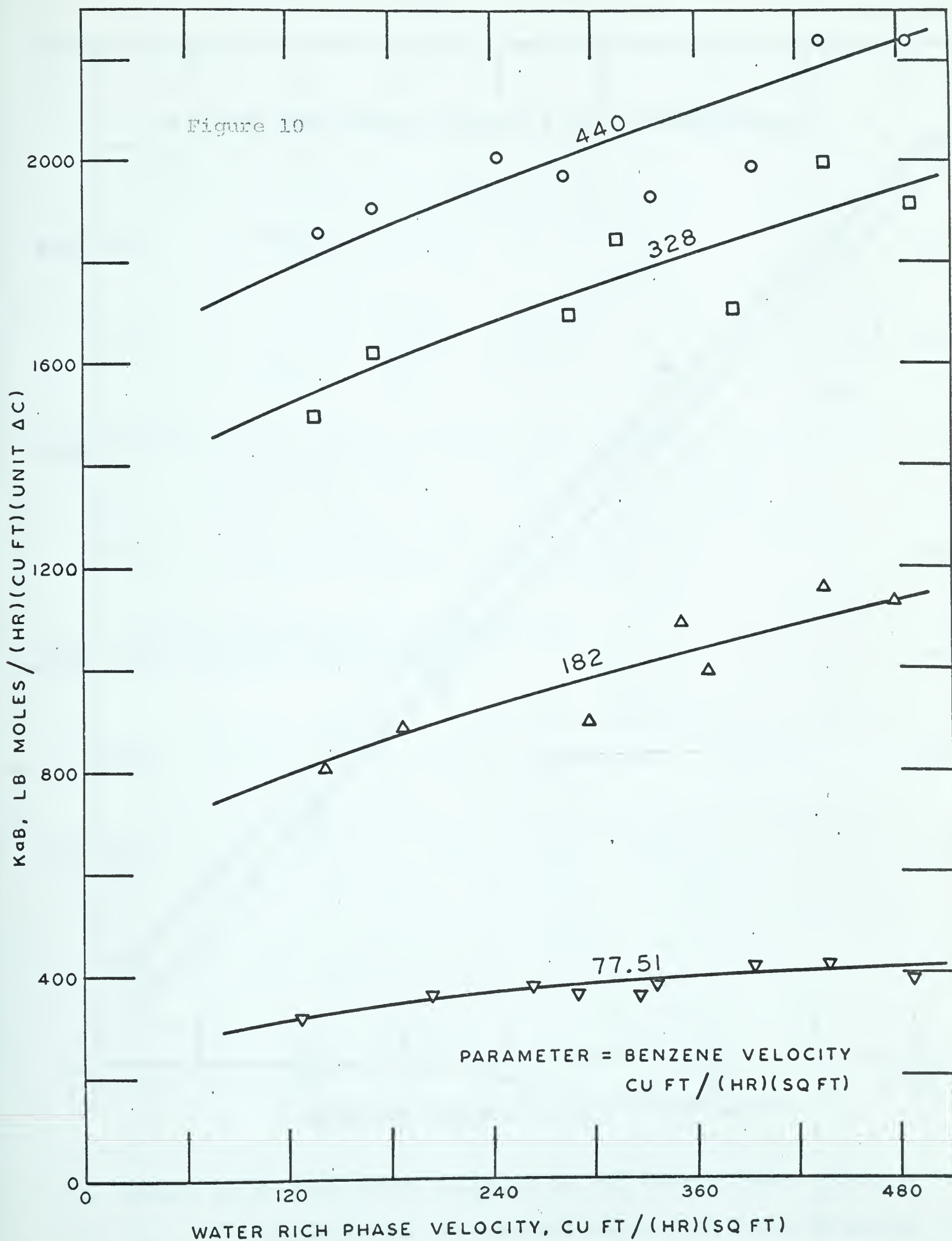
Run numbers 50, 51 and 55, 56 were made using different size settlers with other variables held constant for two consecutive runs. Run numbers 50 and 51 were made with 200 and 500-ml size settlers, respectively. Outlet compositions of benzene-rich phase obtained with the 500-ml settler were only slightly lower than those obtained with the 200-ml settler despite the 2-1/2 to 1 ratio of residence times and approximately 2 to 1 ratio of settling areas. Run numbers 55 and 56 were made with 300 and 500-ml size settlers, respectively. In these runs the outlet benzene-rich phase composition from the 500-ml size settler was slightly higher than the 300-ml size settler. Thus, any effect of settler size is not detectable from these results. The compositions at the settler outlet were therefore treated as the column outlet compositions.

2.4 Effect of Phase Velocities on Overall Mass Transfer Coefficients

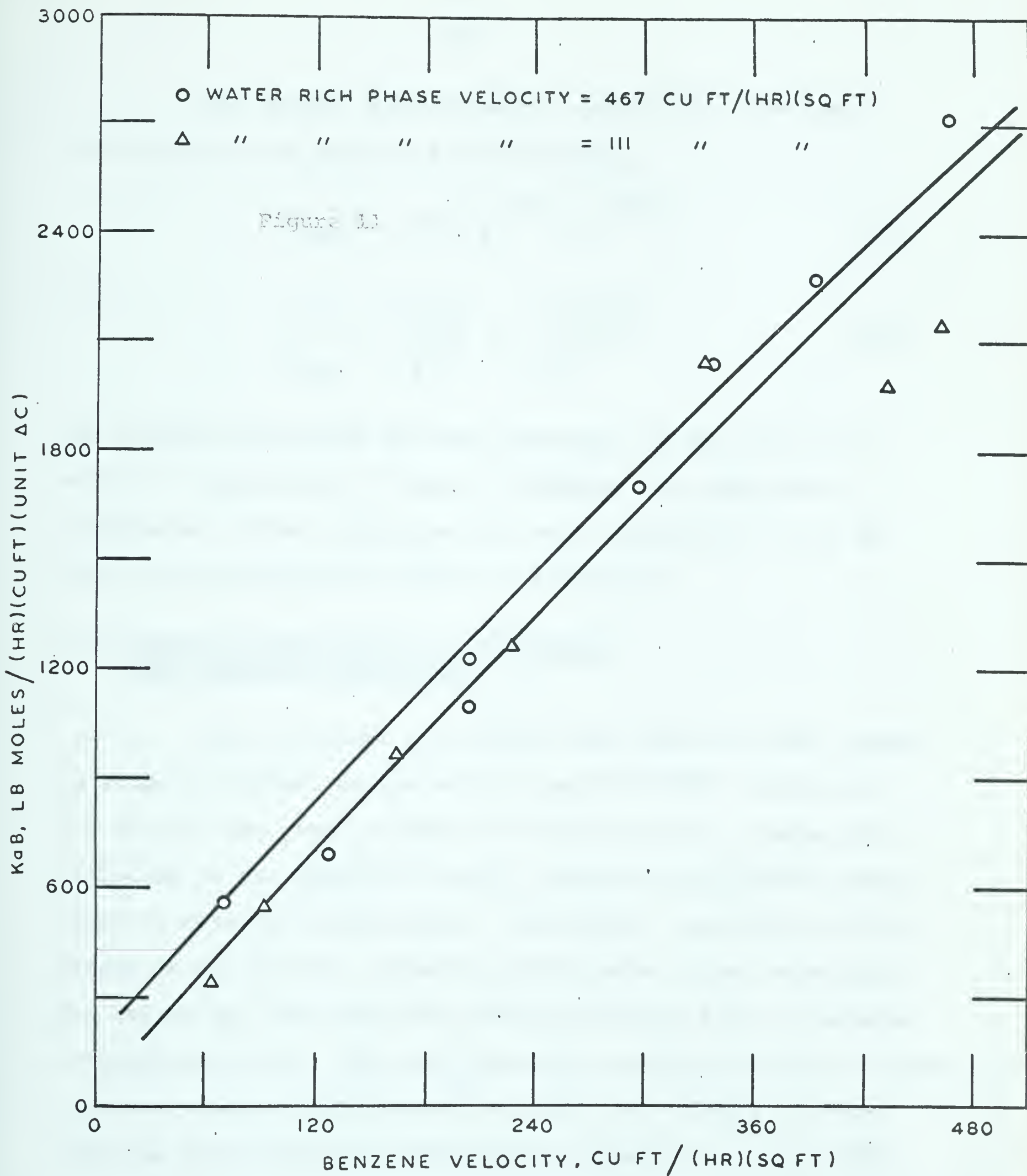
The effect of water-rich phase velocity on the overall mass transfer coefficient at constant benzene velocities is shown in Figure 10. The benzene phase velocity has a pronounced effect on the overall mass transfer coefficients. The scatter in data increases at higher phase velocities because at high velocities outlet compositions are close to the equilibrium value and a slight error in the analysis of acetic acid will have a large effect on the value of overall mass transfer coefficients. The overall mass transfer coefficient increases only slightly with water-rich phase velocity but the effect is greater at higher benzene velocities. At higher benzene phase velocities the benzene phase resistance decreases appreciably and thus one would expect the water-rich phase velocity to have a greater effect on the overall mass transfer coefficient. This is confirmed by the experimental results shown in Figure 10, where the lines have a greater slope at higher benzene velocities.

Figure 11 shows the effect of benzene phase velocity on the overall mass transfer coefficient at constant water-rich phase velocities. The results are in agreement with the above conclusions. The processed data showing the effect of phase velocities is given in Tables III and IV.

Figure 10



Effect of Water-Rich Phase Velocity on the Overall Mass Transfer Coefficient, at Constant Benzene Phase Velocity



Effect of Benzene Phase Velocity on the Overall Mass Transfer Coefficient, at Constant Water-Rich Phase Velocity

The overall mass transfer coefficients have been correlated by the following relationship:

$$K_{aB} = 4.67 V_B^{0.97} V_W^{0.051} \quad (24)$$

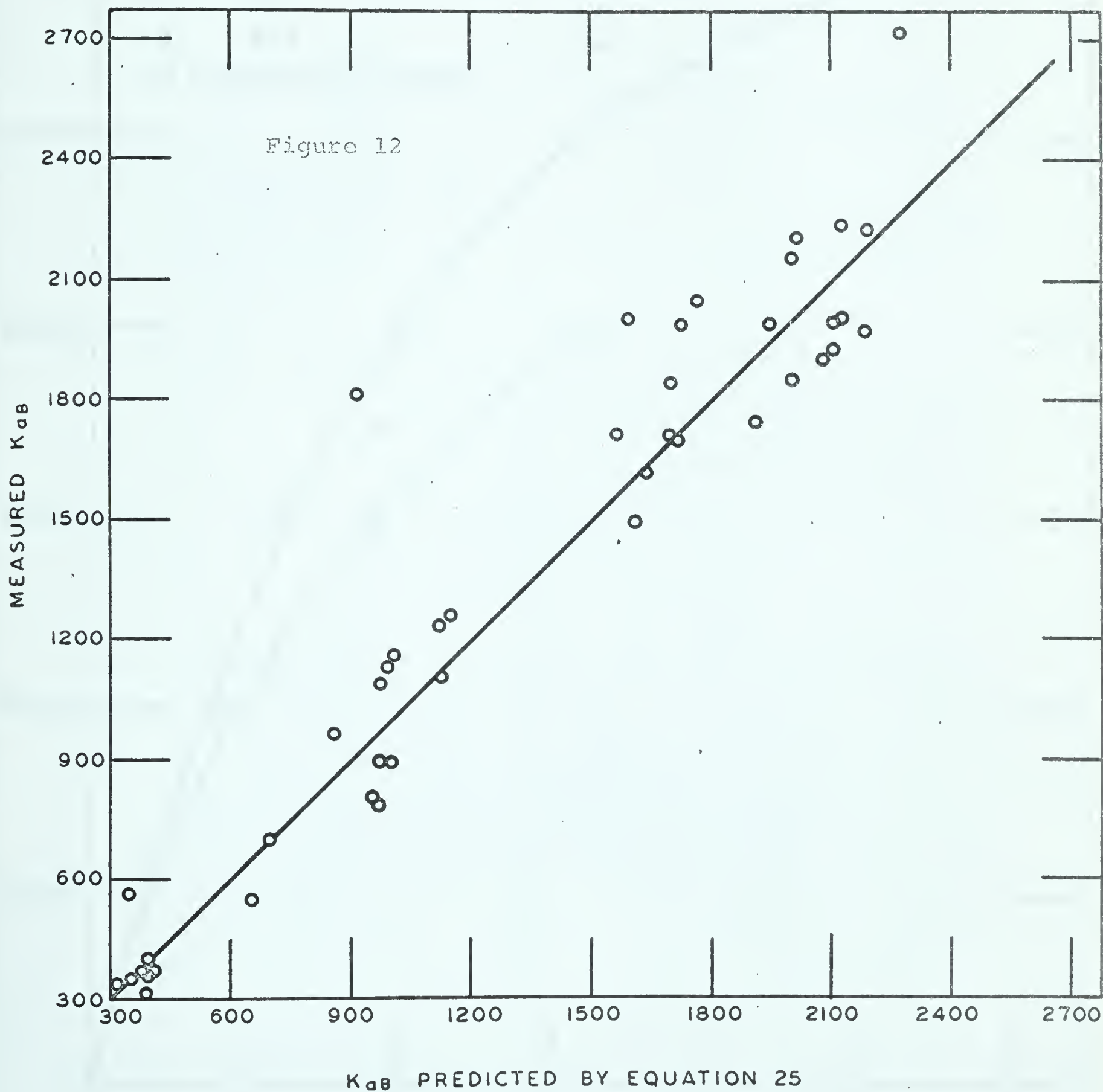
or

$$\frac{1}{K_{aB}} = \frac{0.431}{V_B^{1.2}} + \frac{0.000565}{V_W^{0.2}} \quad (25)$$

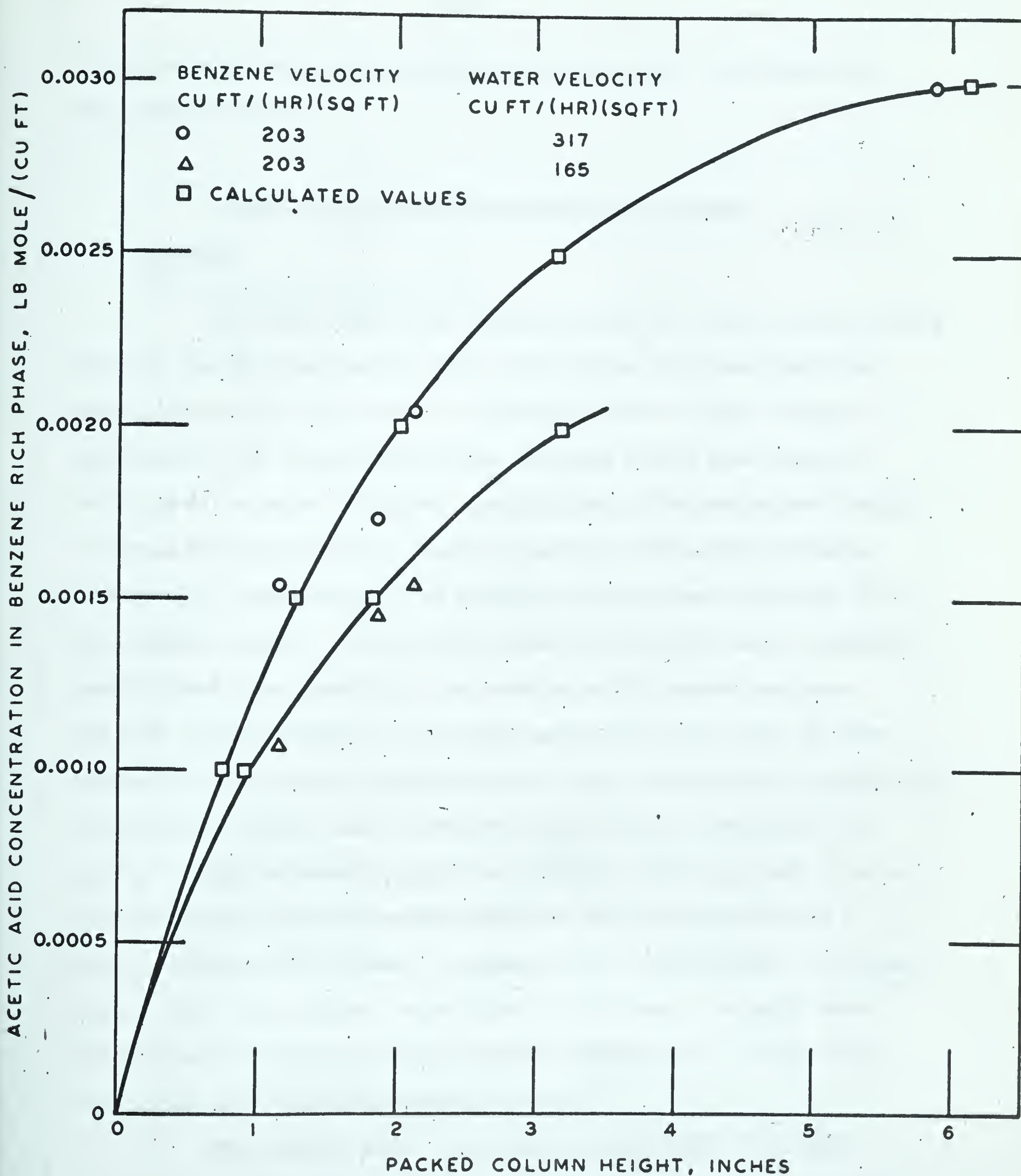
The standard deviation by using equations 24 and 25 is 225 and 227, respectively. Figure 12 compares the experimental results and values calculated by using equation 25. The detail of calculations are given in Appendix C.

2.5 Effect of Bed Height on the Overall Mass Transfer Coefficient

The bed height was varied from 1.153 to 5.890 inches to study its effect on the overall mass transfer coefficient. The results are given in Table II and they show a random distribution in the value of overall transfer coefficients determined at different bed heights. Therefore, any effect of bed height on the overall transfer coefficients is not detectable. The bed height was calculated using equation 23 for an assumed outlet composition. The mass transfer coefficient was calculated from the proposed correlation, equation 24. Figure 13 shows that the calculated and experimental values are in reasonable agreement. This implies that the assumptions made in using a logarithmic mean driving force were reasonable and that there



Comparison of Predicted and Observed Overall Mass Transfer Coefficients in Benzene-Acetic Acid-Water System



Comparison of Observed and Calculated Values of
Acetic Acid Concentration in Benzene-rich Phase

is no appreciable mass transfer at the inlet or outlet of the packed column.

3. Ethyl Acetate-Acetic Acid-Water System

3.1 General

In this study the concentration of acetic acid varied from 26 to 30 lb.mole/(cu.ft.) and Figure 4 shows that the equilibrium line is almost a straight line in this range. Therefore, the logarithmic mean driving force was used to calculate the mass transfer coefficient. The water and ethyl acetate-rich phases were kept saturated with ethyl acetate and water, respectively, to minimize their mass transfer in the packed column. Hence, the calculations for mass transfer coefficient were based on the acetic acid concentrations. Settler outlet compositions were assumed to be equal to the packed column outlet compositions. The distribution coefficient varies with acetic acid concentration and the magnitude of c_W/c_E is approximately equal to 0.87(6), where c_W and c_E are the concentrations of acetic acid in the water-rich and ethyl acetate-rich phases, respectively. Therefore, one would expect that both phase velocities would have a significant effect on the overall mass transfer coefficient. This was confirmed by the experimental results.

The outlet flow rates were calculated from the solubility diagram by using the lever arm rule. The results

indicated that the outlet flow rates were approximately the same as the inlet flow rates. Thus, the inlet flow rates were used for calculating the overall mass transfer coefficients. The material balance for all runs checked within ± 10 percent.

3.2 Effect of Phase Velocities on the Overall Mass Transfer Coefficient

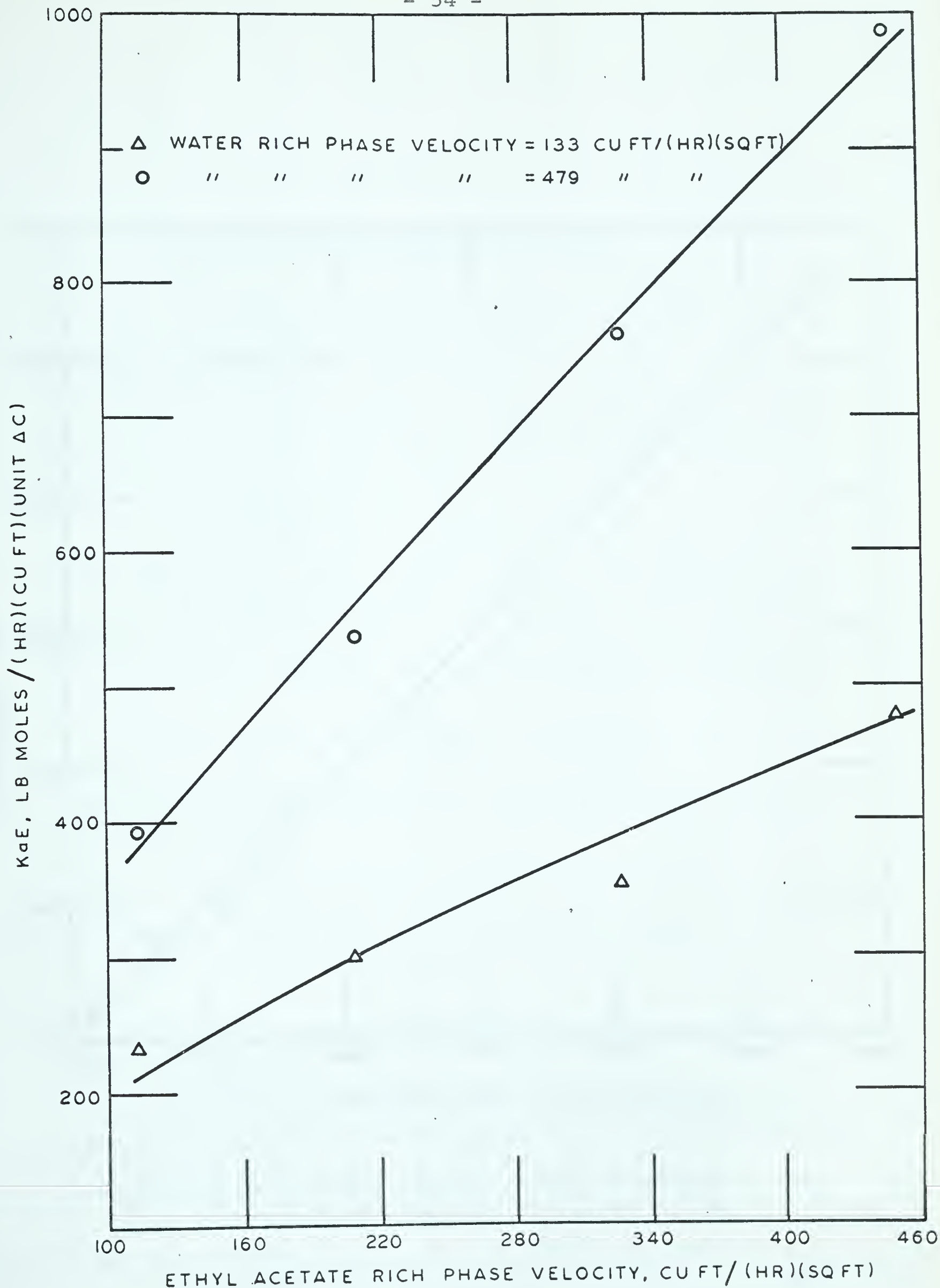
Figure 14 shows the effect of water-rich phase velocity on the overall mass transfer coefficient at constant ethyl acetate-rich phase velocity. The overall transfer coefficient increases with increasing water-rich and ethyl acetate-rich phase velocities and it is equally affected by both phase velocities. The effect of ethyl acetate-rich phase velocity on the overall mass transfer coefficient, at constant water-rich phase velocity, is shown in Figure 15. These results are in agreement with the above conclusions. The processed data showing the effect of phase velocities on the overall mass transfer coefficient are given in Table V.

The overall mass transfer coefficient has been correlated by the following relationship:

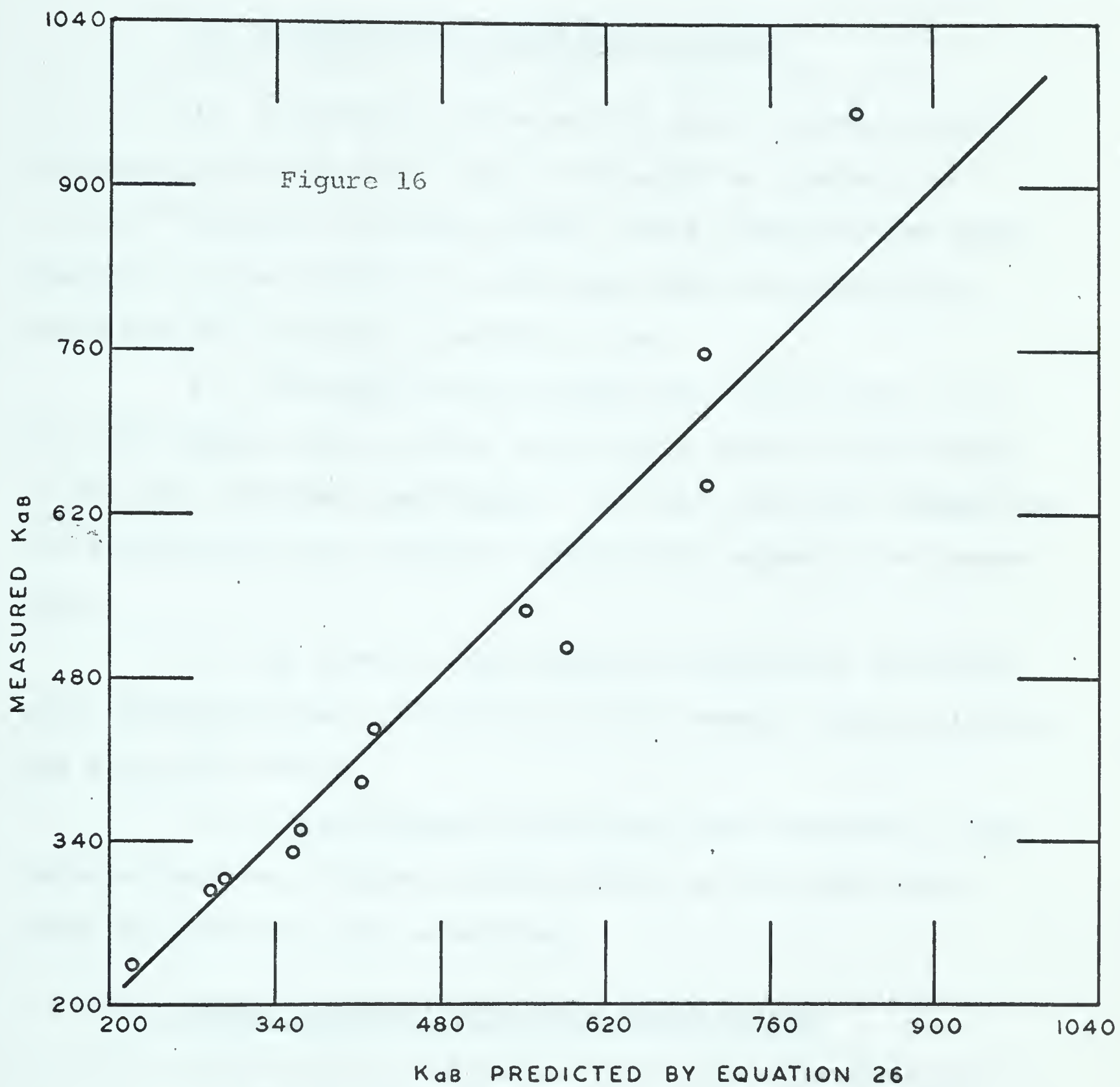
$$K_{aE} = 1.53 V_E^{0.50} V_W^{0.52} \quad (26)$$

The standard deviation by using the above correlation is 63. Figure 16 compares the experimental results and the values calculated by using equation 26. The detail of calculations is given in Appendix C.

Effect of Water-Rich Phase Velocity on the Overall Mass Transfer Coefficient, at Constant Ethyl Acetate-Rich Phase Velocity



Effect of Ethyl Acetate-Rich Phase Velocity on the Overall
Mass Transfer Coefficient, at Constant Water-Rich
Phase Velocity



Comparison of Observed and Predicted Overall Mass Transfer Coefficients in Ethyl Acetate-Acetic Acid-Water System

VI. CONCLUSIONS

1. Benzene-Acetic Acid-Water System

1. In cocurrent flow overall mass transfer coefficients up to 2700 (hr)^{-1} can be obtained as compared to 31 (hr)^{-1} obtained by other workers using countercurrent flow. However, it must be kept in mind that only one equilibrium stage can be attained in cocurrent flow.

2. The packed column height was varied from 1.153 to 5.890 inches and it had no appreciable effect on the value of the mass transfer coefficient. In this study the assumptions for calculating mass transfer coefficients appear to be reasonable.

3. The overall mass transfer coefficient increased with increasing phase velocities and the benzene phase velocity had a greater effect.

4. At high benzene velocities the resistance of the benzene phase was reduced and the effect of the water-rich phase velocity was more pronounced.

2. Ethyl Acetate-Acetic Acid-Water System

1. In cocurrent flow overall mass transfer coefficients up to 960 (hr)^{-1} can be obtained as compared to 6 (hr)^{-1} obtained by other workers using countercurrent flow.

2. The overall mass transfer coefficients increased with increasing phase velocities and were almost equally affected by the water-rich and ethyl acetate-rich phase velocities.

VII. RECOMMENDATIONS FOR FUTURE STUDY

1. In cocurrent flow through packed beds the pressure drop provides the necessary energy needed for mixing and dispersion of the two phases. Thus, it might be feasible to replace the mixer in a mixer settler unit with a packed bed. It would be interesting to make an economic study of the above proposal.
2. Different flow regimes in cocurrent flow have been observed by various workers(12,16,17). It would be interesting to study the mass transfer rate in these flow regimes.
3. Other variables, affecting the overall mass transfer coefficient, which could be studied are the effect of shape and size of packing and column diameter.
4. The data for the ethyl acetate-acetic acid-water system might be extended.

NOMENCLATURE

a	=	area of interfacial contact, sq.ft./(cu.ft. of packed column)
A	=	cross sectional area of packed column, sq.ft.
c	=	solute concentration in the main body of liquid, lb.mole/(cu.ft.)
c_E	=	concentration of transferring component in the extract phase, lb.mole/(cu.ft.)
c_R	=	concentration of transferring component in the raffinate phase, lb.mole/(cu.ft.)
c_O	=	concentration of acetic acid in the organic-rich phase, lb.mole/(cu.ft.)
$(\Delta c)_{1.m}$	=	logarithmic mean terminal potential
d	=	characteristic dimension of the apparatus
d_{32}	=	Sauter mean droplet diameter
D	=	diffusivity, sq.ft./hr.
H	=	liquid holdup
k	=	individual mass transfer coefficient, lb.mole/(hr)(sq.ft.)(unit Δc)
K	=	overall mass transfer coefficient, lb.mole/(hr)(sq.ft.)(unit Δc)
K_a	=	overall mass transfer coefficient on volume basis, lb.mole/(hr)(cu.ft.)(unit Δc)
L	=	liquid flow rate, cu.ft./hr.
m	=	distribution coefficient
N	=	mass transfer rate, lb.mole/(hr)(sq.ft.)
P_{ratio}	=	ratio of frictional pressure gradient to that predicted by means of the "single phase" assumption

ΔP	=	pressure drop, inches of water
S	=	interfacial area, sq.ft.
u	=	liquid velocity in z direction, cm/sec.
V	=	superficial velocity, cu.ft./(hr)(sq.ft. of column cross section)
y	=	distance from the gas liquid interface
z	=	packed bed height, ft.

Greek Symbols

μ	=	viscosity, lb/(hr)(ft)
∇	=	gas liquid interfacial tension
ρ	=	density, lb/(cu.ft.)

Subscripts

B	=	referring to benzene-rich phase
C	=	referring to continuous phase
D	=	referring to dispersed phase
E	=	referring to ethyl acetate-rich and extract phase
o	=	referring to organic-rich phase
R	=	referring to raffinate phase
W	=	referring to water-rich phase

BIBLIOGRAPHY

1. Comings, E.W., and Briggs, S.W., "Liquid-Liquid Extraction in Wetted Wall and Packed Columns", Trans A.I.Ch.E., 38, 143 (1942).
2. Davis, J.T., and Rideal, E.K., "Spontaneous Emulsification" in "Interfacial Phenomena", Academic Press, New York, (1961).
3. Dodds, W.S., Stuteman, L.F., Sollami, B.J., and McCarter, R.J., "Concurrent Gas Absorption Mass Transfer", A.I.Ch.E.J. 6, 197 (1960).
4. Dodds, W.S., Stutzman, L.F., Sollami, B.J., and McCarter, R.J., "Pressure Drop and Liquid Holdup in Concurrent Gas Absorption", A.I.Ch.E.J., 6, 391 (1960).
5. Eaglesfield, P., Kelly, B.K. and Short, J.F., "Extraction of Acetic Acid from Dilute Solutions", Industrial Chemistry, 29, 147 and 243 (1953).
6. Finzi, S., Renzoni, R., Silvistri, M., and Villani, S., "Mass and Heat Transfer in Cocurrent Liquid-Gas Flow", Energia Nucleare, 4, 211 (1957).
7. Garner, F.G., Ellis, S.R.M., and Roy, V.N.G., "Extraction of Acetic Acid from Water 1. Benzene-Acetic Acid-Water", Chem. Eng., Sci., 2, 14 (1953).
8. Garner, F.G., and Ellis, S.R.M., "Extraction of Acetic Acid from Water 2. Acetic Acid-Water-Ethyl Acetate", Chem. Eng. Sci., 2, 282 (1953).
9. Groothuis, H., and Zuiderweg, F.J., "Influence of Mass Transfer on Coalescence of Drops", Chem. Eng. Sci., 12, 288 (1960).
10. Leacock, J.A., and Churchill, S.W., "Mass Transfer Between Isobutanol and Water in Concurrent Flow Through Packed Beds", A.I.Ch.E.J., 7, 196 (1961).
11. Rigg, R.G., and Churchill, S.W., "The Behavior of Immiscible Liquids in Cocurrent Flow Through Packed Beds", A.I.Ch.E.J., 10, 811 (1964).
12. Scott, D.S., "Properties of Cocurrent Gas-Liquid Flow" in "Advances in Chemical Engineering", Vol. 4, Academic Press, New York (1963).

13. Scott, D.S. and Hayduk, W., "Gas Absorption in Horizontal Cocurrent Bubble Flow", Paper presented at Oct., 1964 meeting of Chemical Institute of Canada.
14. Sherwood, T.K., Evans, J.E., and Longson, J.V.A., "Extraction in Spray and Packed Columns", Trans. A.I.Ch.E., 35, 597, (1939).
15. Tang, Y.P. and Himmelblau, D.M., "Interphase Mass Transfer for Laminar Cocurrent Flow of Carbon Dioxide and Water Between Parallel Plates", A.I.Ch.E.J., 9, 630 (1963).
16. Weekman, V.W., and Myers, J.E., "Fluid Flow Characteristics of Concurrent Gas-Liquid Flow in Packed Beds", A.I.Ch.E.J., 10, 951 (1964).
17. Wen, C.Y., O'Brien, W.S. and Fan, L., "Pressure Drop Through Packed Beds Operated Cocurrently", J. of Chem. & Eng. Data, 8, 47 (1963).
18. Wen, C.Y., O'Brien, W.S. and Fan, L., "Mass Transfer in Packed Beds Operated Cocurrently", J. of Chem. & Eng. Data, 8, 42 (1963).

A P P E N D I X A
TABLES OF PROCESSED DATA

The following symbols are used in this Appendix:

- V_W = Water-rich phase velocity, cu.ft./((hr)(sq.ft.))
- V_B = Benzene rich phase velocity, cu.ft./((hr)(sq.ft.))
- V_E = Ethyl acetate-rich phase velocity, cu.ft./((hr)(sq.ft.))
- c_W = Acetic acid concentration in the water-rich phase
- c_B = Acetic acid concentration in the benzene-rich phase
- c_E = Acetic acid concentration in the ethyl acetate-rich phase
- K_{aB} = Overall mass transfer coefficient based on concentrations in the benzene-rich phase, lb.moles/((hr)(cu.ft.)) (unit Δc)
- K_{aE} = Overall mass transfer coefficient based on concentrations in the ethyl acetate-rich phase, lb.moles/((hr)(cu.ft.)) (unit Δc)
- H = Height of the packed column, inches
- D = Average diameter of the packed column, inches

Subscripts

- 1 = concentration at the inlet of packed column
- 2 = concentration at the exit of packed column

1. MASS TRANSFER DATA

SYSTEM: BENZENE-ACETIC ACID-WATER

TABLE II

Effect of Packed Column Height on the Mass Transfer Coefficient

Run Number	V_W	V_B	C_{W1}	C_{W2}	C_{B1}	C_{B2}	K_{AB}	H	D
1	163.9	207.8	81.4×10^{-3}	79.9×10^{-3}	0	1.07×10^{-3}	879	1.153	0.588
2	311.1	206.6	81.4	80.2	0	1.54	1424	1.153	0.588
3	163.4	211.3	81.4	79.0	0	1.45	839	1.862	0.583
4	316.4	210.3	81.4	79.9	0	1.73	1061	1.862	0.583
5	165.1	212.4	81.4	79.0	0	1.54	808	2.129	0.580
6	319.7	212.4	81.4	79.6	0	2.05	1244	2.129	0.580
7	171.5	184.4	74.4	73.8	0	2.47	868	5.890	0.575
8	321.8	180.9	81.4	79.3	0	2.99	1088	5.890	0.575

TABLE III

Effect of Water-Rich Phase Velocity on the Mass Transfer Coefficient

Run Number	V_W	V_B	C_{W1}	C_{W2}	C_{B1}	C_{B2}	K_{aB}	H	D
9	129.2	77.5	90.8×10^{-3}	89.6×10^{-3}	0	3.30×10^{-3}	316	5.890	0.575
10	203.2	77.5	90.8	90.2	0	3.46	360	5.890	0.575
11	261.9	77.5	90.8	90.5	0	3.52	378	5.890	0.575
12	294.9	74.0	90.8	90.8	0	3.52	356	5.890	0.575
13	327.6	77.5	90.8	90.5	0	3.49	357	5.890	0.575
14	335.8	77.5	90.8	90.2	0	3.49	374	5.890	0.575
15	392.7	77.5	90.8	90.5	0	3.59	411	5.890	0.575
16	436.9	77.5	90.8	90.5	0	3.59	416	5.890	0.575
17	488.5	77.5	90.8	90.8	0	3.55	388	5.890	0.575
18	142.1	182.0	76.4	73.6	0	2.45	805	5.890	0.575
19	184.4	182.0	76.4	74.3	0	2.54	881	5.890	0.575
20	293.6	180.9	76.4	74.9	0	2.56	899	5.890	0.575
21	321.8	180.9	81.4	79.3	0	2.99	1088	5.890	0.575
22	366.4	180.9	76.4	75.2	0	2.58	895	5.890	0.575
23	435.7	184.4	81.4	79.6	0	2.99	1159	5.890	0.575

TABLE III (continued)

Run Number	V_W	V_B	c_{W1}	c_{W2}	c_{B1}	c_{B2}	K_{aB}	H	D
24	475.6	182.03	76.4×10^{-3}	75.5×10^{-3}	0	2.64×10^{-3}	1130	5.890	0.575
25	136.2	328.8	76.4	71.5	0	2.35	1495	5.890	0.575
26	171.5	328.8	76.4	72.1	0	2.43	1620	5.890	0.575
27	286.5	328.8	76.4	73.7	0	2.52	1691	5.890	0.575
28	314.7	327.6	81.4	78.4	0	2.89	1845	5.890	0.575
29	381.7	327.6	76.4	74.3	0	2.56	1705	5.890	0.575
30	435.7	328.8	81.4	79.0	0	2.94	1993	5.890	0.575
31	486.2	330.0	76.4	74.7	0	2.63	1913	5.890	0.575
32	138.6	441.6	90.8	82.9	0	3.01	1855	5.890	0.575
33	171.5	447.4	90.8	84.1	0	3.08	1905	5.890	0.575
34	244.3	454.5	90.8	85.9	0	3.21	2057	5.890	0.575
35	283.0	458.0	90.8	86.9	0	3.27	1969	5.890	0.575
36	335.8	432.2	90.8	87.2	0	3.28	1927	5.890	0.575
37	394.6	439.2	90.8	87.8	0	3.35	1989	5.890	0.575
38	434.5	441.6	90.8	88.4	0	3.43	2232	5.890	0.575
39	485.0	451.0	90.8	88.9	0	3.44	2236	5.890	0.575

TABLE IV

Effect of Benzene-Rich Phase Velocity on the Mass Transfer Coefficient

Run Number	V_W	V_B	C_{W1}	C_{W2}	C_{B1}	C_{B2}	K_{aB}	H	D
40	108.9	63.7	72.6×10^{-3}	71.4×10^{-3}	0	2.23×10^{-3}	340	4.935	0.579
41	111.2	122.8	72.6	71.4	0	2.29	552	4.935	0.579
42	111.2	164.5	72.6	70.4	0	2.17	962	4.935	0.579
43	108.9	172.6	72.6	68.7	0	2.20	1814	4.935	0.579
44	111.2	225.8	72.6	69.2	0	2.18	1266	4.935	0.579
45	111.2	333.6	72.6	67.3	0	2.12	2046	4.935	0.579
46	108.9	429.7	72.6	66.1	0	2.08	1984	4.935	0.579
47	108.9	461.0	72.6	66.1	0	2.09	2151	4.935	0.579
48	466.7	69.5	90.1	89.6	0	3.40	559	4.935	0.579
49	471.4	128.6	90.1	88.1	0	3.31	698	4.935	0.579
⁺ 50	466.7	203.8	90.1	86.6	0	3.26	1232	4.935	0.579
*51	466.7	203.8	90.1	86.1	0	3.19	1158	4.935	0.579

TABLE IV (continued)

Run Number	V_W	V_B	C_{W1}	C_{W2}	C_{B1}	C_{B2}	K_{aB}	H	D
52	466.7	295.3	87.4×10^{-3}	84.7×10^{-3}	0	3.13×10^{-3}	1708	4.935	0.579
53	467.9	338.2	87.4	83.2	0	3.10	2036	4.935	0.579
54	466.7	391.5	87.4	83.8	0	3.10	2280	4.935	0.579
*55	466.7	459.8	87.4	82.9	0	3.07	2726	4.935	0.579
x56	464.4	454.4	87.4	83.5	0	3.04	2694	4.935	0.579

* Settler Size = 500 c.c.

+ Settler Size = 200 c.c.

x Settler Size = 300 c.c.

SYSTEM: ETHYL ACETATE-ACETIC ACID-WATER

TABLE V

Effect of Water-Rich and Ethyl Acetate-Rich Phase Velocities on the Mass Transfer Coefficient

Run Number	V _W	V _E	C _{W1}	C _{W2}	C _{E1}	C _{E2}	K _{aE}	H	D
57	477.9	112.7	86.7x10 ⁻³	73.2x10 ⁻³	0.620x10 ⁻³	68.8x10 ⁻³	392	5.890	0.575
58	477.9	206.9	86.7	64.8	0.620	56.8	537	5.890	0.575
59	477.9	330.0	86.7	57.7	0.620	50.0	758	5.890	0.575
60	481.5	445.1	86.7	51.9	0.620	45.7	966	5.890	0.575
61	251.3	116.3	86.7	60.1	0.620	57.7	306	5.890	0.575
62	358.2	112.7	86.7	70.4	0.620	63.0	331	5.890	0.575
63	135.0	112.7	86.7	56.8	0.620	47.2	232	5.890	0.575
64	131.5	210.2	86.7	45.1	0.620	34.3	298	5.890	0.575
65	131.5	326.5	86.7	36.6	0.620	25.2	350	5.890	0.575
66	131.5	449.1	86.7	29.2	0.620	20.4	427	5.890	0.575
67	244.3	448.6	86.7	42.3	0.620	28.4	507	5.890	0.575
68	351.1	448.6	86.7	47.7	0.620	34.9	644	5.890	0.575

2. EQUILIBRIUM DATA

TABLE VI

Equilibrium Data for
Benzene-Acetic Acid-Water System

Serial Number	Acetic Acid in Water- Rich Phase lb.mole/(cu.ft.)	Acetic Acid in Benzene-Rich Phase lb.mole/(cu.ft.)
1	31.66×10^{-3}	0.7261×10^{-3}
2	47.63	1.298
3	47.90	1.253
4	64.08	2.082
5	80.74	3.382
6	96.47	4.105
7	144.1	8.029

A P P E N D I X B
DETAILS OF LEAST SQUARE ANALYSIS

1. Benzene-Acetic Acid-Water System

1.1 Determination of Coefficients

Two correlations for calculating the overall mass transfer coefficients were proposed and the detail of calculations are discussed separately.

(1) The following relationship

$$K_{aB} = a_o V_B^{a_1} V_W^{a_2}$$

may be written as

$$\log K_{aB} = \log a_o + a_1 \log V_B + a_2 \log V_W$$

Let

$$b = \begin{bmatrix} \log a_o \\ a_1 \\ a_2 \end{bmatrix}, \quad y = \begin{bmatrix} \log K_{aB}(1) \\ \log K_{aB}(2) \\ - \\ - \\ - \\ \log K_{aB}(N) \end{bmatrix}, \quad x = \begin{bmatrix} 1.0 & \log V_B(1) & \log V_W(1) \\ 1.0 & \log V_B(2) & \log V_W(2) \\ - & - & - \\ - & - & - \\ 1.0 & \log V_B(N) & \log V_W(N) \end{bmatrix}$$

where y represents the experimental N values of mass transfer coefficients. In the present case N = 47. The least square values of $\log a_o$, a_1 and a_2 were determined by solving the following set of equations by Crout's method.

$$x^T x b = x^T y$$

The calculations were carried out on the IBM 7040 computer in floating point mode and required only 14 seconds of machine time exclusive of input and output.

(2) The following correlation for overall mass transfer coefficient

$$\frac{1}{K_{aB}} = \frac{b_1}{V_B^P} + \frac{b_2}{V_W^Q}$$

may be written as

$$1/K_{aB} = b_1 V_B^{-P} + b_2 V_W^{-Q}$$

One hundred and fifty pairs of P and Q were assumed and the values of b_1 and b_2 were calculated by least square analysis. Let,

$$b = \begin{bmatrix} b_1 \\ b_2 \end{bmatrix}, \quad Y = \begin{bmatrix} 1/K_{aB}(1) \\ 1/K_{aB}(2) \\ - \\ - \\ 1/K_{aB}(N) \end{bmatrix}, \quad X = \begin{bmatrix} V_B(1)^{-P} & V_W(1)^{-Q} \\ V_B(2)^{-P} & V_W(2)^{-Q} \\ - & - \\ - & - \\ V_B(N)^{-P} & V_W(N)^{-Q} \end{bmatrix}$$

where Y represents the experimental N values of mass transfer coefficients. In the present case $N = 47$. The least square values of b_1 and b_2 were determined by solving the following set of equations by the elimination method.

$$X^T X b = X^T Y$$

The variance for each assumed pair of P and Q was calculated. The final value of P and Q was selected corresponding to a minimum variance.

The calculations were carried out on the IBM 7040 computer in floating point mode and required 3 minutes and 13 seconds of machine time exclusive of input and output.

1.2 Determination of Standard Variance

The standard variance is defined as the square root of variance. The variance was calculated from the following formula:

$$\sigma^2 = \frac{\sum_{i=1}^N \left[K_{ai} - \phi_m(x_i) \right]^2}{N - (m + 1)}$$

where N = number of mass transfer coefficient values used for least square calculations

$\phi_m(x)$ = the proposed correlation

$(m + 1)$ = linearly independent functions, equal to 3 and 2 for equation (24) and (25) respectively

2. Ethyl Acetate-Acetic Acid-Water System

The following correlation for overall mass transfer coefficients was proposed:

$$K_{aE} = a_0 V_E^{a_1} V_W^{a_2}$$

The constants a_0 , a_1 and a_2 were calculated exactly in the same manner as for the benzene-acetic acid-water system.

A P P E N D I X C

SAMPLE CALCULATIONS

1. Calculation of Equilibrium Data

Normality of a solution was calculated from the following formula:

$$N_1 V_1 = N_2 V_2$$

where N = normality of the solution

V = volume of the solution used in titration

Subscripts 1 and 2 refer to the solution numbers one and two, respectively.

1.1 Standardization of Sodium Hydroxide Solution

A known weight of oxalic acid was dissolved in distilled water and the volume of solution was made up to 100 c.c. Equivalent weight of oxalic acid is equal to 63.035.

TABLE VII

Calculation of Sodium Hydroxide Normality

	Solution Number One	Solution Number Two
Weight of oxalic acid dissolved	6.2556 gm	6.3020 gm
Normality of oxalic acid solution	$\frac{6.2556}{63.035} \times \frac{1}{10}$ = 0.9926 N	$\frac{6.3020}{63.035} \times \frac{1}{10}$ = 0.9998 N
volume of oxalic acid used	10 cc	10 cc
volume of sodium hydroxide solution used	10.05 cc	10.35 cc
Normality of sodium hydroxide solution	$\frac{0.9926 \times 10}{10.05}$ = 0.9637 N	$\frac{0.9998 \times 10}{10.35}$ = 0.9657 N

Arithmetic mean of sodium hydroxide normalities

$$= \frac{0.9678 + 0.9637}{2} = 0.9657$$

Percentage difference in the two values of sodium hydroxide

$$\text{normalities} = \frac{0.9768 - 0.9637}{0.9657} \times 100 = 0.423$$

Similarly normality of 1/40 - N sodium hydroxide was calculated which was equal to 0.02417.

1.2 Equilibrium Data Calculations

The water-rich and benzene-rich phases were titrated with 1 - N and 1/40 - N standard sodium hydroxide solutions respectively.

Volume of sample used = 10 cc

TABLE VIII

Calculation of Water-Rich and
Benzene-Rich Phases Normality

	Water-Rich Phase	Benzene-Rich Phase
Volume of sodium hydroxide solution used	7.9 cc	8.6 cc
Normality of samples	$\frac{0.9657 \times 7.9}{10}$ = 0.7629 N	$\frac{0.02417 \times 8.6}{10}$ = 0.02055 N
lb mole of acetic acid in the samples per cu.ft.	$\frac{0.7629 \times 28.32}{453.59}$ = 0.04763	$\frac{0.02055 \times 28.32}{453.59}$ = 0.01298

2. Calculation of Mass Transfer Coefficient

Concentration of benzene-rich and water-rich phases were calculated exactly in the same way as described in the previous section.

Height of the packed column, z = 1.153 inches

Average diameter of the packed

column, D = 0.565 inch

Inlet concentration of

benzene-rich phase, c_{B1} = 0.0 lb.mole/cu.ft.

Outlet concentration of

benzene-rich phase, c_{B2} = 0.001067 lb.mole/cu.ft.

Inlet concentration of

water-rich phase, c_{W1} = 0.08141 lb.mole/cu.ft.

Outlet concentration of

water-rich phase, c_{W2} = 0.0799 lb.mole/cu.ft.

From Figure 9 concentration of benzene-rich phase

(i) at the entrance of the packed column,

c_{Be1} = 0.00342 lb.mole/cu.ft., and

(ii) at the outlet of the packed column,

c_{Be2} = 0.00314 lb.mole/cu.ft.

Cross sectional area of the

packed column = $\pi D^2/4$

$$= \frac{\pi \times (0.565)^2}{4}$$

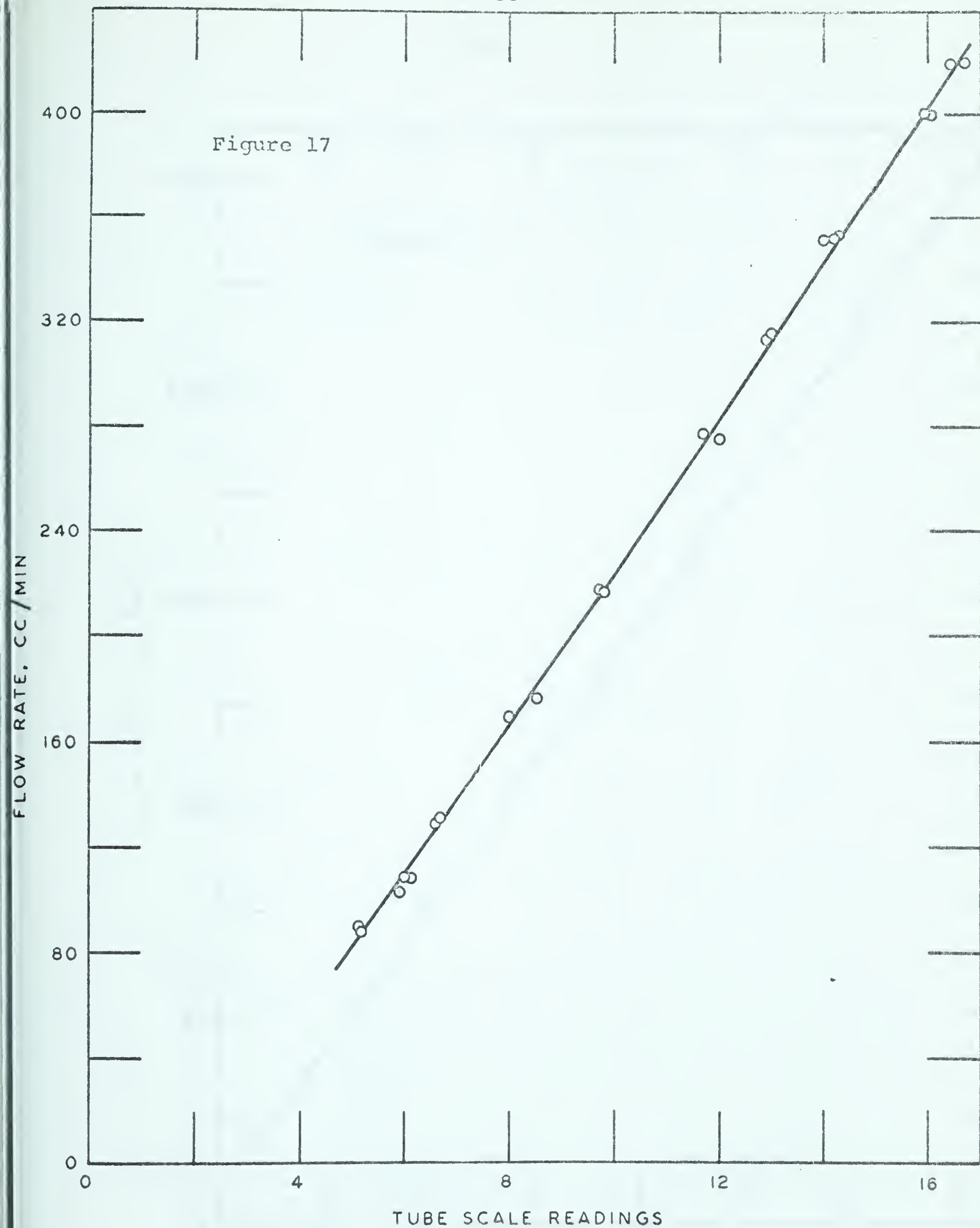
= 0.00174 sq.ft.

Therefore benzene-rich phase velocity,

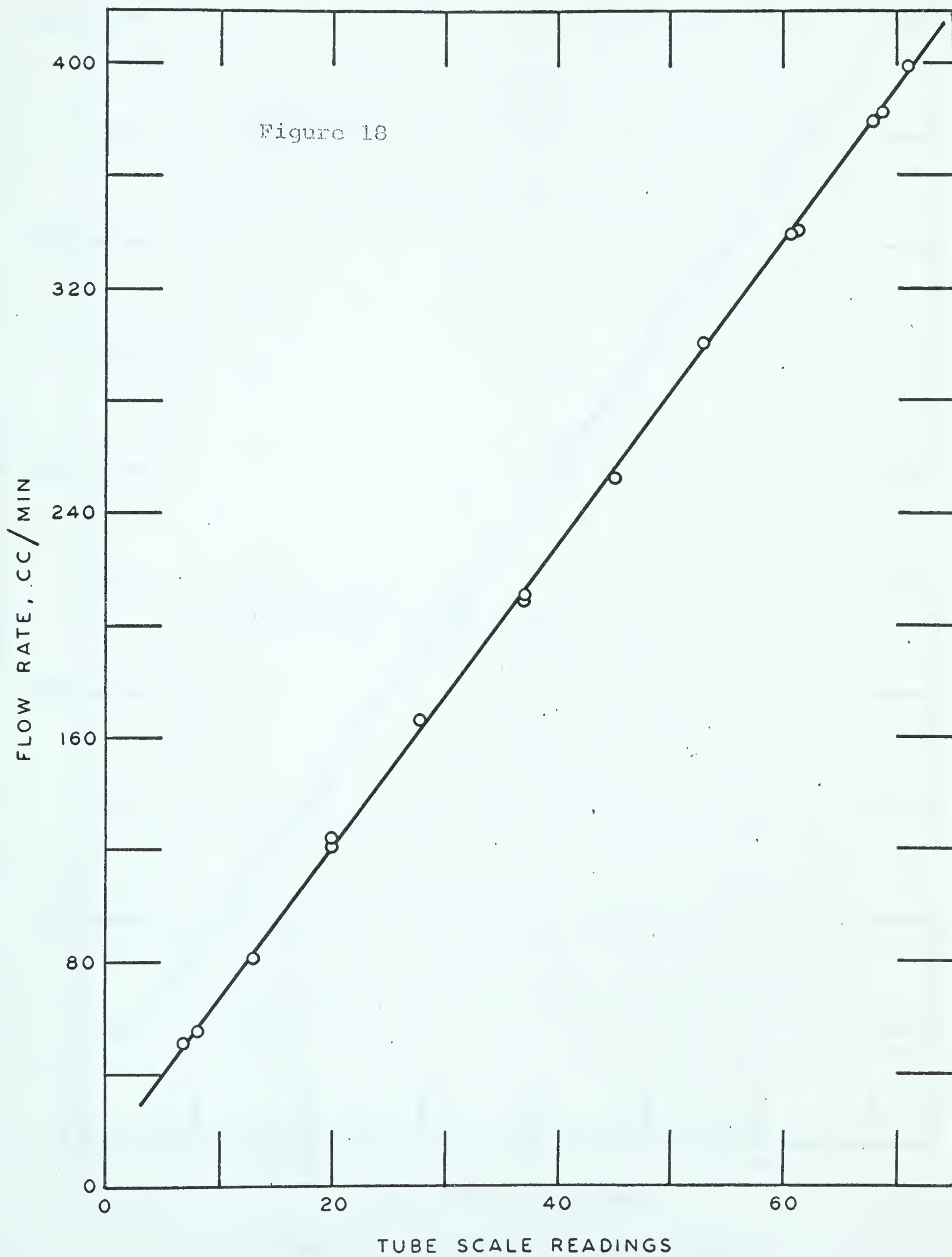
$$\begin{aligned}
 V_B &= 185 \times 0.00003531 \times 60 \text{ cu.ft./hr} \\
 &= \frac{185 \times 0.00003531 \times 60}{0.00174} \text{ cu.ft./hr(sq.ft.)} \\
 &= 207.76 \\
 (\Delta c_B)_{l.m} &= \frac{(c_{Be} - c_B)_1 - (c_{Be} - c_B)_2}{\ln \left[\frac{(c_{Be} - c_B)_1}{(c_{Be} - c_B)_2} \right]} \\
 &= \frac{(0.00342 - 0.0) - (0.00314 - 0.001067)}{2.303 \log \left(\frac{0.00342 - 0.0}{0.00314 - 0.001067} \right)} \\
 &= 0.002736 \\
 K_{aB} &= \frac{V_B (c_{B2} - c_{B1})}{z (\Delta c_B)_{l.m}} \\
 &= \frac{207.76 \times 0.001067 \times 12}{1.153 \times 0.002736} \\
 &= 842 \text{ lb.mole/((hr)(cu.ft.)(}\Delta c\text{)}
 \end{aligned}$$

The calculations for overall mass transfer coefficients in ethyl acetate-acetic acid-water system were carried out exactly in the same manner.

A P P E N D I X D
CALIBRATION OF ROTAMETERS

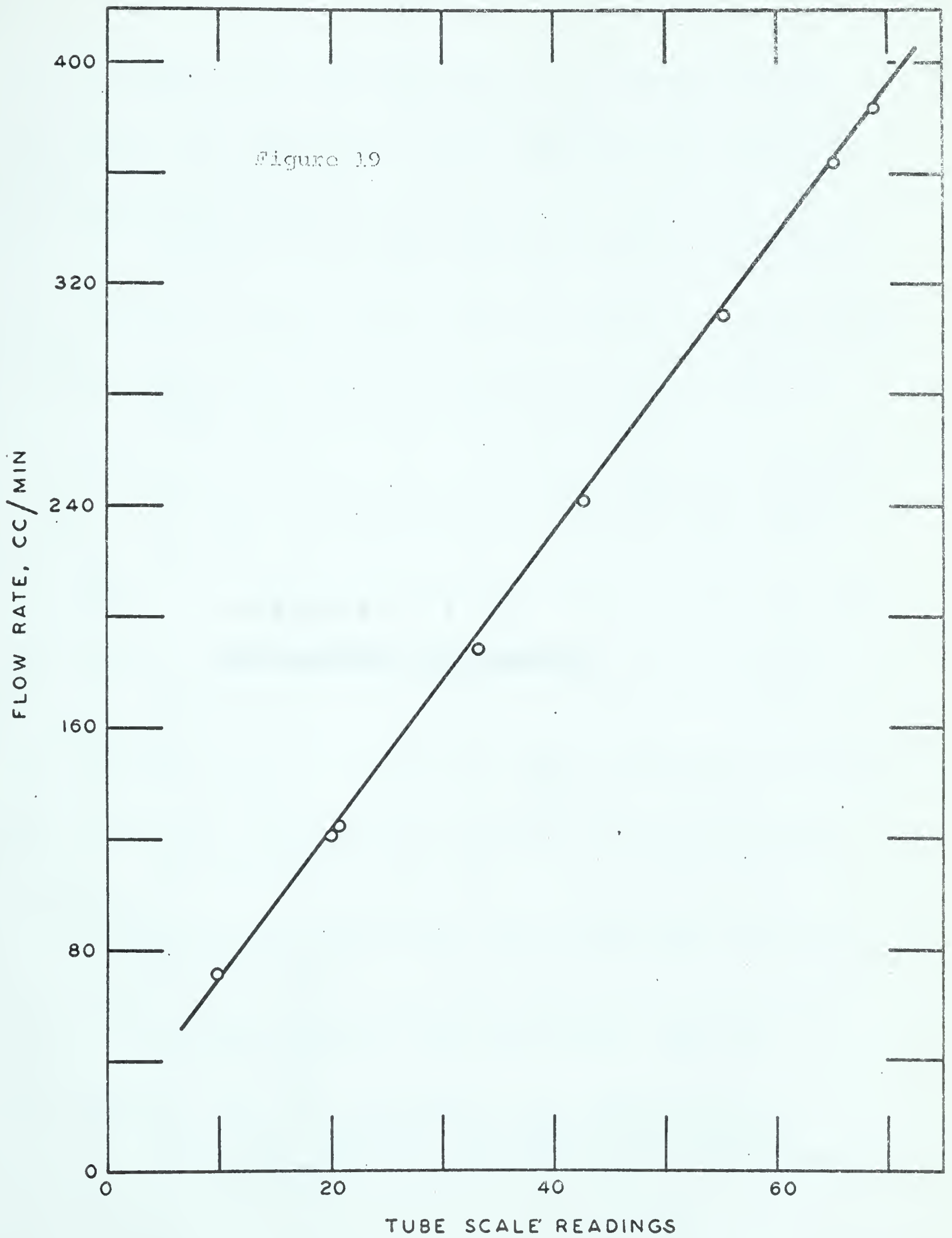


Rotameter Calibration, Liquid: Aqueous Solution of Acetic Acid



Rotameter Calibration, Liquid:

Ethyl Acetate - Saturated with Water



Rotameter Calibration, Liquid. Benzene

A P P E N D I X E
SUPPLEMENTARY BIBLIOGRAPHY

1. Bischoff, K.B., and Himmelblau, D.M., "Survey of Mass Transfer", Ind. Eng. Chem., 56, 61 (Dec., 1964).
2. Brain, W.A., and Hougen, O.A., "Flooding Velocities in Packed Columns", Trans. A.I.Ch.E., 40, 29 (1944).
3. Christensen, G.B., and Terjesen, S.G., "On the Action of Surface Active Agents on Mass Transfer in Liquid-Liquid Extraction", Chem. Eng. Sci., 9, 225 (1959).
4. Danckwerts, P.V., "Significance of Liquid-Film Coefficients in Gas Absorption", Ind. Eng. Chem., 43, 1460 (1951).
5. Davidson, J.F., "Application of Penetration Theory For Mass Transfer in Packed Columns", Trans. Inst. Chem. Eng. (London), 37, 131 (1959).
6. Galloway, L.R., Komarnicky, W. and Epstein, N., "Effect of Packing Configuration on Mass and Heat Transfer in Beds of Stacked Spheres", Can. J. Chem. Eng., 35, 139 (Dec. 1957).
7. Hills, S., "Channeling in Packed Columns", Chem. Eng. Sci., 1, 247 (1952).
8. Jackson, M.L., Holman, K.L., and Grove, D.B., "Surface Effects in Pulsed, Packed Column", A.I.Ch.E.J., 8, 659 (1962).
9. Jeffreys, G.V., "Efficiency of Liquid Extraction Processes", Chem. & Eng. Process Eng., 43, 117 (1962).
10. Lewis, J.B., "The Mechanism of Mass Transfer of Solute Across Liquid-Liquid Interfaces", Chem. Eng. Sci., 8, 295 (1958).
11. Liles, A.W., and Geankophs, C.J., "Axial Diffusion of Liquids in Packed Beds and End Effects", A.I.Ch.E.J., 6, 591 (1960).
12. Miller, D.N., "Liquid Film Controlled Mass Transfer in Agitated Vessels", Ind. Eng. Chem., 56, 19 (Oct., 1964).
13. Moorhead, D.H. and Himmelblau, D.M., "Optimization of Operating Variables in a Packed Liquid-Liquid Extraction Column", Ind. & Eng. Chem. Fundamentals, 1, 68 (1962).

14. Othmer, D.F., "Acetic Acid Recovery Methods", Chem. Eng. Prog., 54, 48 (1958).
15. Rao, M.R., and Rao, C.V., "Flooding Rates in Packed Liquid Extraction Tower", Chem. Eng. Sci., 9, 170 (1958).
16. Sakiadis, B.C., and Johnson, A.F., "Generalized Correlation of Flooding Rates", Ind. Eng. Chem., 46, 1229 (1954).
17. Shulman, H.L., and Margolis, J.E., "Performance of Packed Columns", A.I.Ch.E.J., 3, 157 (1957).
18. Smoot, L.D. and Babb, A.L., "Mass Transfer Studies in a Pulsed Extraction Column", Ind. & Eng. Chem. Fundamentals, 1, 92 (1962).
19. Spalding, D.B., "Prediction of Mass Transfer Rates When Equilibrium Does Not Prevail at Phase Interface", Inst. J. Heat & Mass Transfer, 2, 283 (1961).
20. Venkatraman, G. and Laddha, G.S., "Limiting Velocities, Holdup and Pressure Drop in Packed Extraction Columns", A.I.Ch.E.J., 6, 355 (1960).
21. Victor, M.S. and Poffenberger, N., "Commercial Extraction Equipment", Ind. Eng. Chem., 42, 1021 (1950).
22. Victor, M.S., and Beckman, R.B., "Temperature Studies in Packed Liquid-Liquid Extraction Tower", Ind. Eng. Chem., 42, 1078 (1950).
23. Wilke, C.R., Prausnitz, J.M., Acrivos, A., Petersen, E.E., and Olander, D.R., "Mass Transfer", Ind. Eng. Chem., 53, 925 (1961).

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